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Controlling magnetic and transport properties of granular alloys through Joule heating

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Melt-spun Cu–Co ribbons are annealed by linearly varying current Joule heating. During the annealing, the electrical resistance is measured in order to follow the structural transformations within the samples. The resistance versus current curves show a characteristic behavior for all samples studied. This fact is used to specify optimum conditions to obtain the best nanostructure which displays the maximum giant magnetoresistance ratio. © *1998 American Institute of Physics.* [S0021-8979(98)04521-6]

Giant magnetoresistance (GMR) has been observed in granular Cu–Co alloys[1–3] and subsequently in a number of other granular metals, where ferromagnetic particles are embedded in a nonmagnetic matrix.[4,5] It is generally accepted that the mechanism responsible for this effect is the spin-dependent electron scattering within the grains and at their boundaries.[6] In most experiments the samples are annealed to modify the nanostructure, and a maximum of GMR is observed in the annealing temperature range of 500–550 °C, as a consequence of the precipitation, growth, and coarsening of the magnetic particles.[7–11] What is still lacking, however, is an effective procedure to control such structural transformations during the annealing process.

In Joule heating (JH) techniques, an electrical current is applied to anneal metallic samples, releasing heat and consequently increasing the temperature of the material. These techniques have been successfully used to anneal amorphous metallic materials and granular solids, resulting in better magnetic and mechanical properties.[9,12–14] Owing to the possibility of obtaining high heating and cooling rates ($\approx 100\ \mathrm{K\,s^{-1}}$), pulses of high current are usually applied (also known as flash annealing), which allows one to obtain off-equilibrium phases. However, this "fast" Joule heating hinders the possibility of monitoring the structural transformations which take place in the samples. This drawback can be overcome by slowly increasing the applied current and following the behavior of the electrical resistance R during annealing, a technique introduced here and called linearly varying current Joule heating (LVC-JH).

In this communication, we apply LVC-JH to anneal $Cu_{0.90}Co_{0.10}$ and $Cu_{0.85}Co_{0.15}$ melt-spun ribbons. We assert that it is possible to control the structural transformations which occur during annealing by the on-line monitoring of the sample's resistance. An accurate control can be performed because the resistance behavior is extremely reproducible in all samples studied, independent of fluctuations on the sample's width, thickness and initial resistance. The annealing current I was interrupted at different points of the *R(I)* characteristic curve, and structural, magnetic and transport properties were investigated to achieve an optimum structure which presents the best GMR ratio.

The experimental setup of the LVC-JH is composed of two pairs of U-shaped contacts used for clamping the ends of the sample within a small region in order to minimize thermal losses by conduction. The current I, applied by two of the contacts, is varied step-by-step (increment $\Delta I = 0.1$ A, time interval $\Delta t = 10$ s), from 0 to 15 A. The heating rate $\Delta I/\Delta t$ can be properly adjusted according to the dynamics of the processes observed during the treatment. The voltage drop V across the sample is measured through the other two contacts, giving a direct measurement of the electrical resistance $R = V/I$. During the annealing, the sample is kept in a 5×10^{-2} mbar vacuum in order to avoid oxidation and minimize convective thermal losses. The measurement is completely computer controlled, and therefore the on-line analysis of both $R(I)$ and $dR(I)/dI$ curves allows one to precisely determine at which point the annealing can be interrupted. It is also possible to specify the way the annealing current is decreased to zero: slow decrease or abrupt interruption.

The LVC-JH was applied in 0.1 m long ribbon strips of pure Cu, $Cu_{0.90}Co_{0.10}$, and $Cu_{0.85}Co_{0.15}$ (average width $l = 5$ mm and thickness $d = 50\ \mu$m) which were obtained by planar flow casting in vacuum. Figure 1 shows the characteristic $R(I)$ curves. For pure Cu, the experimental data is fitted using the JH model for metallic ribbons presented in Ref. 13. In this model, the Joule power RI^2 is responsible for the heating of the sample, while radiation is the main heat loss mechanism. In alloys which display a positive temperature coefficient of resistance (TCR), an increase of temperature gives rise to an increase of resistance, and the expected curve is a parabola such as the one obtained for pure Cu [see Fig. 1(c)].[13] The same model properly fits the Cu–Co curves for small currents ($I < 5$ A for $Cu_{0.90}Co_{0.10}$, and $I < 3.5$ A for $Cu_{0.85}Co_{0.15}$, see Fig. 1). For higher values of I, the resistance drops as a consequence of the Co precipitation, and a clear maximum is observed. As the grain size enlarges, the interparticle separation increases and structural disorder less-

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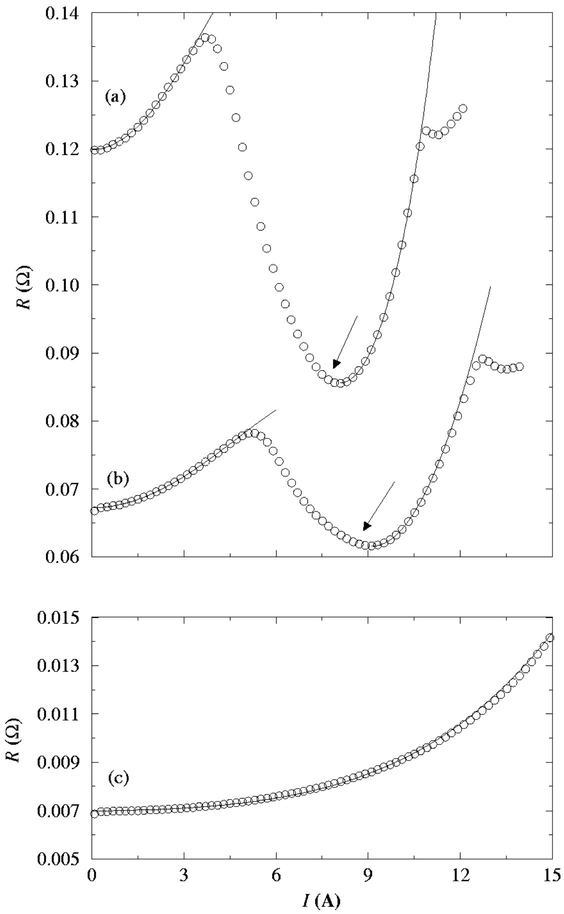

FIG. 1. Typical resistance R vs electrical current I curves for (a) $Cu_{0.85}Co_{0.15}$, (b) $Cu_{0.90}Co_{0.10}$, and (c) pure Cu. Arrows indicate the points where the largest GMR ratios were found.

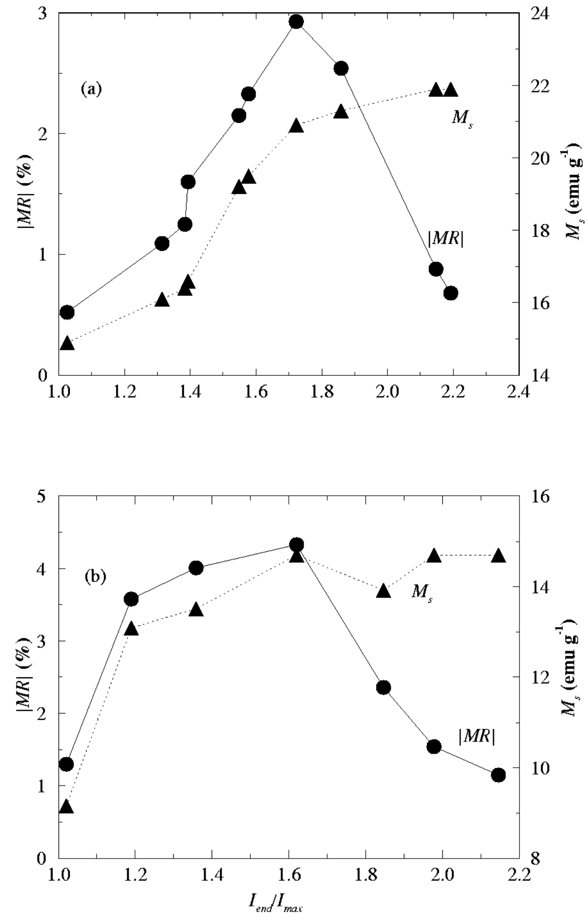

FIG. 2. GMR ratio (circles) and saturation magnetization (triangles) as functions of the current at which the Joule heating was stopped $I_{\rm end}$, normalized by the value of current where the first maximum in the $R(I)$ curves occurs $I_{\rm max}$: (a) $Cu_{0.85}Co_{0.15}$ and (b) $Cu_{0.90}Co_{0.10}$.

ens, resulting in the decrease of resistance, down to a minimum. After this minimum, the samples seem to behave again like an ordinary metal, until another transformation (for $I > 11$ A), probably related to the coalescence of the precipitates,[15] takes place. All samples studied (more than 100) have displayed a similar behavior, independent of their initial resistance, width, and thickness. Actually, for each composition, the $R(I)$ curves closely follow a universal behavior, if one properly normalizes the current and resistance axes by the corresponding values at the first maximum ($I_{\rm max}$, $R_{\rm max}$).

Using the characteristic $R(I)$ curve as a guide, we abruptly interrupted the LVC-JH at different current values, in order to follow the structural transformations which occur in these samples. Besides Cu $K_{\alpha 1}$ x-ray diffractometry, we have indirectly determined the structural and magnetic parameters of the samples through magnetic data. The magnetization and magnetoresistance were measured at room temperature, using a vibrating sample magnetometer with the magnetic field H varying from 0 to 1.4 T.

In the initial stages of Co precipitation, the magnetic grains are small enough to behave as superparamagnetic particles, and the magnetization curves are accurately fitted by a superposition of Langevin functions weighted by a log-normal distribution function. In these cases it is possible to obtain the magnetic moment distribution and the fraction of precipitated Co that contribute to the saturation magnetization.[16]

Figure 2 shows the saturation magnetization M_s and GMR ratio $[{\rm MR}=100\times(R_{H=1.4\,{\rm T}}-R_{H=0\,{\rm T}})/R_{H=0\,{\rm T}}]$ for two sets of samples obtained by stopping the treatment at a certain current $I_{\rm end}$ [$Cu_{0.85}Co_{0.15}$, Fig. 2(a) and $Cu_{0.90}Co_{0.10}$, Fig. 2(b)]. In both plots $I_{\rm end}$ is normalized by the values of the current at a first maximum ($I_{\rm max}$) of the corresponding $R(I)$ curve. Before the maximum, the saturation magnetization remains almost constant (≈ 9 emu/g for $Cu_{0.90}Co_{0.10}$ and ≈ 15 emu/g for $Cu_{0.85}Co_{0.15}$, not shown in Fig. 2). After the first maximum, M_s gradually increases, achieving the limit value (21.9 emu/g for $Cu_{0.85}Co_{0.15}$ and 15.1 emu/g for $Cu_{0.90}Co_{0.10}$) when all Co present in the sample precipitates. From an analysis of the magnetic data, we infer that the evolution of the magnetically active Co fraction probably starts at the inflection point before the first maximum of the $R(I)$ curve and continues just until the minimum, at which practically all the Co contributes to the saturation magnetization. The largest GMR ratio occurs for $I_{\rm end}$ close to the minimum in the characteristic $R(I)$ curve (see the arrows indicating the corresponding points in Fig. 1). In these points, the estimated mean Co grain size is 1.8 nm for $Cu_{0.85}Co_{0.15}$ and 1.1 nm for $Cu_{0.90}Co_{0.10}$. After the minimum

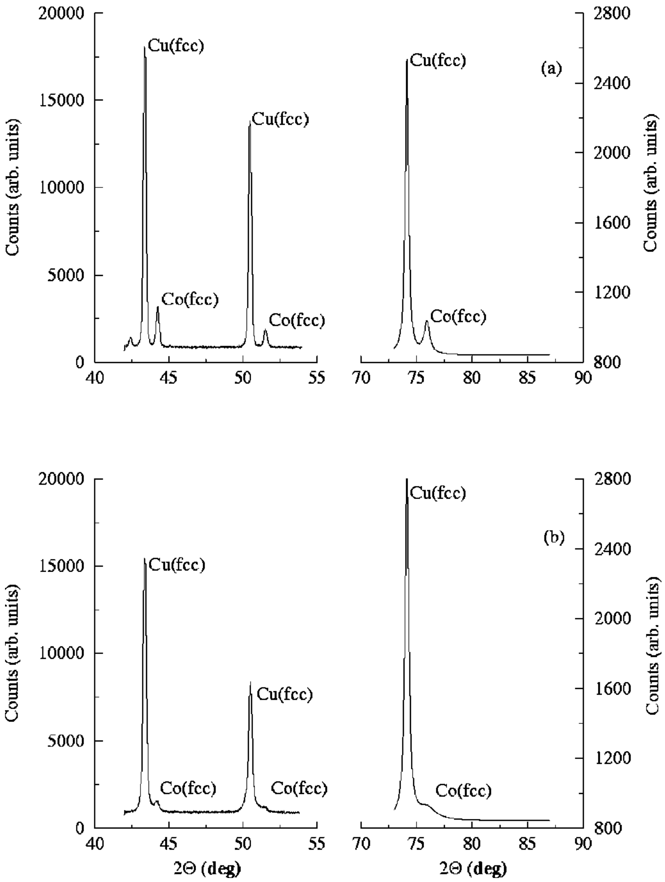

FIG. 3. X-ray diffraction data of the same samples shown in Fig. 1: (a) $Cu_{0.85}Co_{0.15}$ and (b) $Cu_{0.90}Co_{0.10}$.

in $R(I)$ curves, the magnetization curves of all annealed samples cannot be fitted by the superparamagnetic model, and it is observed that the GMR ratio starts decreasing. This effect is attributed to the growth and coarsening of blocked magnetic grains. In fact, for high values of I_{end} [after the second maximum of the $R(I)$ curve] the x-ray diffraction profiles shown in Fig. 3 reveal the fcc structure of the already large Co grains. The fcc structure was observed independently of the way the electrical current was decreased.

The evolution of the sample's resistance as a function of the annealing current is successfully used to monitor the Co precipitation. Generally speaking, all $R(I)$ curves display a similar behavior, a fact that can be explored to stop the annealing at a convenient point. The results indicate that the maximum of the GMR ratio occurs when almost all the Co is magnetically active, but the grains are still superparamagnetic—more precisely, at $I_{end}/I_{max}=1.62$ for $Cu_{0.90}Co_{0.10}$ and at $I_{end}/I_{max}=1.72$ for $Cu_{0.85}Co_{0.15}$. Furthermore, it is worth noting that the experimental procedure is rapid, feasible and extremely reproducible, enabling a precise determination of the point where the material will display the best GMR properties.

In conclusion, we employed the LVC variation of the Joule heating technique to follow the structural (precipitation and growth of Co grains) and magnetic (saturation magnetization and GMR ratio) properties of granular Cu–Co alloys. The results indicate that it is therefore possible to establish a precise criterium to perform optimal thermal treatments. The reproducibility of the experimental results suggests that some other properties related to the structure of the material, especially those of technological interest, can be controlled by the same method.

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