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Magnetic and magnetotransport properties in Joule-heated granular $Cu_{95}Co_5$ ribbons

Paolo Allia [a], Marco Coïsson [b,*], Paola Tiberto [b], Franco Vinai [b]

[a] Politecnico di Torino, Dipartimento di Fisica, I-10129 Torino, Italy

[b] IEN Galileo Ferraris, Materials Department, Strada delle Cacce 91, I-10135 Torino, Italy

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Abstract

$Cu_{95}Co_5$ alloys, consisting of Co nanoparticles, were obtained by subjecting rapidly quenched ribbons to suitable dc Joule-heating treatments ($0 \leq I \leq 5.75$ A for 60 s) in order to induce different degrees of Co precipitation with particles having different average size. The behaviour of the resistance versus applied field is influenced by the structural changes (Co cluster formation) that occur in the samples after different thermal treatments. The dependance of the electric and magnetic properties on microstructural configurations has been studied by measuring room-temperature hysteresis loops and GMR. By combining the GMR response with magnetization data, information on the magnetic correlation between magnetic moments of neighbouring Co particles will be obtained and correlated to the different microstructures induced by current annealing.

Keywords: Nanostructured materials; Magnetoresistance; Rapid solidification

1. Introduction

Melt-spun granular alloys [1], consisting of nanosized magnetic particles dispersed in a non-magnetic matrix, have been extensively investigated due to the presence of a significant resistance variation under an applied magnetic field (giant magneto-resistance or GMR) [2,3]. During the last decade, it progressively became clearer that the conventional superparamagnetic (SP) model is an excessively crude picture of the magnetic and magnetotransport properties of these materials. Systematic studies have evidenced that in melt-spun granular alloys it is very common to invoke an improved model that takes into account the presence of dipolar interactions among magnetic nanoparticles dispersed in the non-magnetic matrix (so-called ISP regime) [4]. The search for alloys displaying pure or almost pure SP behaviour led sometimes to efforts in decreasing the content of the magnetic component in the alloy.

In this paper, a rapidly solidified $Cu_{95}Co_5$ (nominal composition) alloy has been produced and selected samples have been subjected to non-conventional current annealing (Joule heating) in order to favour the precipitation of nanosized magnetic clusters from the out of equilibrium solid solution of Co in Cu. While always being in its SP state, the alloy properties still show a remarkable dependence on the annealing current, leading to improved magnetic and magnetoresistance properties for an annealing current above 5 A.

2. Experimental

A continuous ribbon of nominal composition $Cu_{95}Co_5$ has been produced by melt-spinning in vacuum. Selected samples have been subjected to dc Joule heating in vacuum with current intensities ranging in the interval $0 \leq I \leq 5.75$ A for $t = 60$, in order to induce different degrees of Co precipitation with particles having different average size. Room-temperature hysteresis loops were obtained by using an Alternating Gradient Force Magnetometer under a field up to 20 kOe. Magnetoresistance measurements were performed at room temperature with the conventional four-contacts technique under an applied field up to 20 kOe (field and current in sample plane, but orthogonal to each other).

3. Results and discussion

Magnetization curves as a function of magnetic field on as quenched and current annealed samples never display any detectable hysteresis; the shape of the $M(H)$ curve can always be fitted with a superposition of Langevin functions where the Co particle moments and their relative weights are left as a free fitting parameter [5]. The quality of the fitting is always very good.

* Corresponding author. Tel.: +39 011 3919 855; fax: +39 011 3919 834.
E-mail address: coisson@ien.it (M. Coïsson).

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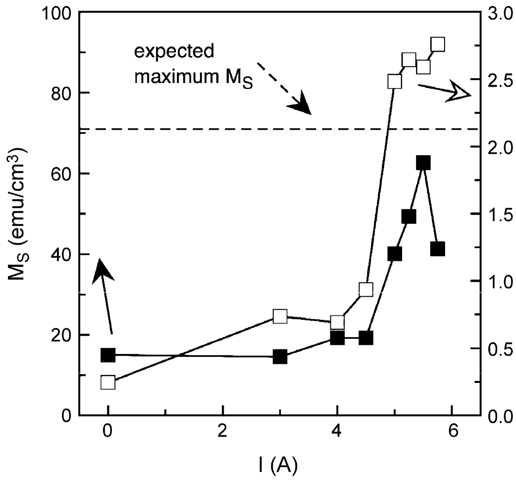

Fig. 1. Magnetic saturation (full symbols) and maximum GMR value (open symbols) as a function of annealing current intensity. The dashed line is the expected magnetic saturation value if all the Co atoms were grouped into particles contributing to the magnetization of the alloy.

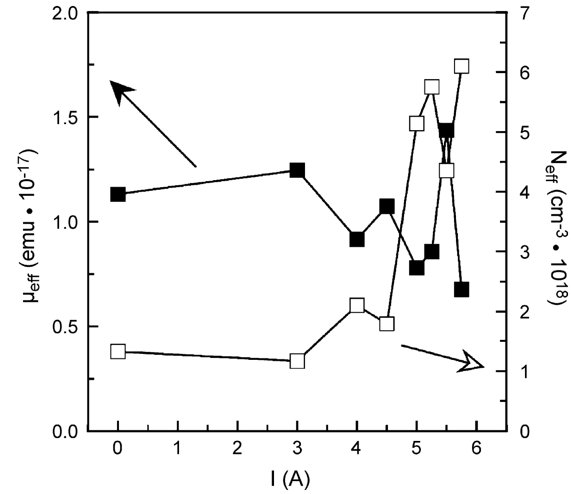

Fig. 2. Effective magnetic moment (full symbols) and effective number (open symbols) of Co particles dispersed in the alloy.

According to the ISP model [4], the *effective* magnetic moment μ_{eff} and diameter D_{eff} of the Co particles can be obtained with this analysis, as well as the effective number of particles N_{eff} per unit volume; their effective average distance d_{eff} is simply equal to $N_{\mathrm{eff}}^{-1/3}$.

The magnetic saturation value of all studied samples is reported in full symbols in Fig. 1. The dashed line is the magnetic saturation value that the alloy should display if all the Co atoms were segregated into particles contributing to the magnetization of the alloy. In the as quenched conditions, the alloy saturation is clearly very far away from its maximum theoretical value, indicating that a large fraction of Co atoms are dispersed into the Cu matrix as a result of the rapid solidification process (it is usually expected that about 3 at.%. of Co can be dispersed in Cu with this technique [6]). Joule heating enhances Co atoms precipitation into particles [7,8] for heating currents above 4.5 A; when $I = 5.5$ A the alloy saturation reaches a maximum, while the reduced M_S value of the sample annealed at $I = 5.75$ A possibly indicates the beginning of Co redissolution in the Cu matrix. Fig. 1 also reports as open symbols the maximum GMR value of the studied samples as a function of the annealing current; strongly annealed samples display the best GMR response, indicating that the current induced Co precipitation favours the presence of small and numerous particles instead of the growth of few larger ones.

Fig. 2 reports as full symbols the effective magnetic moment associated to the average Co particle as a function of annealing current, as results from fitting procedure described above. The samples displaying the largest GMR values also display the lowest μ_{eff} values, indicating that the annealing process enhances the formation of small Co particles responsible of the GMR effect. At $I = 5.5$ A, μ_{eff} first abruptly increases, indicating that small Co particles start to coalesce into larger magnetic units (detrimental to GMR), then higher current intensities act to decrease the effective moment per particle, marking the onset of Co redissolution into the Cu matrix, according to the behaviour of the saturation magnetization reported in Fig. 1. Fig. 2 also shows as open symbols the effective number of Co particles, as a function of annealing current. A higher particle density corresponds to smaller particles, but when small particles start to coalesce, their number per unit volume decreases, possibly indicating that larger Co particles may actually be an effective cluster of smaller, structurally independent but magnetically interacting Co particles. For the highest annealing current value the final increase of N_{eff} may indicate the loss of magnetic correlation among the small particles that formed the larger clusters, thus resulting in a larger number of independent Co particles in the alloy (as will be explained later in the comment of Fig. 4).

Fig. 3 displays the effective average distance d_{eff} among Co particles (full squares). Samples annealed with currents ≥5 A are clearly characterized by smaller (see Fig. 2) and nearer Co particles, whose relative distance slightly increases for the sample annealed at $I = 5.5$ A that displays the larger (and thus more separated) clusters made of interacting smaller Co particles. The effective diameter over effective distance ratio is also reported

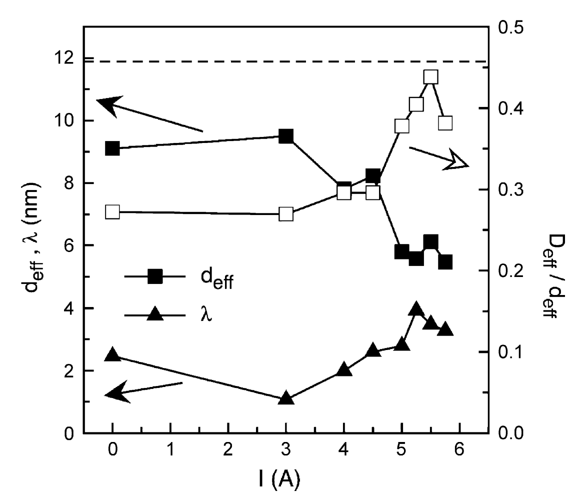

Fig. 3. Full squares: effective distance between Co particles dispersed in the alloy. Open squares: ratio of the effective diameter of Co particles and their effective distance (dashed line: geometrical prediction). Full triangles: room temperature conduction electron mean free path.

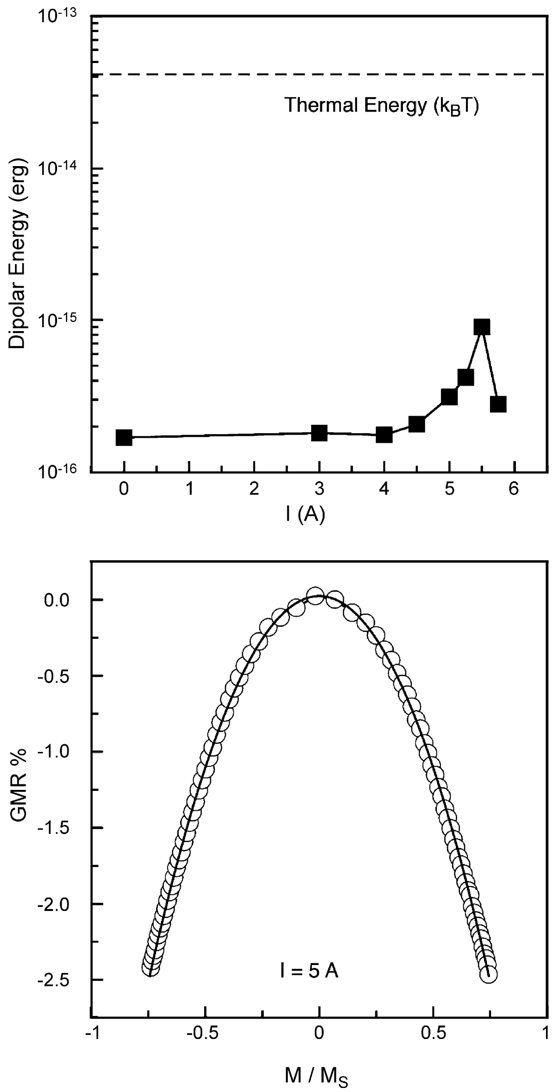

Fig. 4. Top: dipolar energy of interaction among Co particles dispersed in the alloy, as a function of annealing current; the dashed line is the room temperature thermal energy. Bottom: GMR vs. reduced magnetization curve for the sample annealed with $I = 5$ A (open symbols); the line is the parabolic best fit of the experimental data.

as open squares; the initially low values of this ratio indicate that the as quenched sample and those annealed at $I < 5$ A are characterized by few particles, while most of the Co atoms are dispersed in the Cu matrix. On the contrary, the sample annealed at $I = 5.5$ A is characterized by almost all Co atoms forming clusters, since the D_{eff}/d_{eff} ratio is very close to its predicted value for Co spheres in a 5 at.% Co alloy. The possible redissolution of Co into the Cu matrix at the highest current intensities observed in Fig. 1 is confirmed by the corresponding reduction of the D_{eff}/d_{eff} ratio. Fig. 3 also shows the conduction electron mean free path λ as a function of annealing current as calculated from resistivity measurements using the Drude formula. λ is always smaller than d_{eff}; those samples displaying the largest GMR ratio are characterized by λ values closer to d_{eff}, thus maximizing the probability of magnetic scattering of the conduction electron on Co particle surfaces.

The fact that λ is always smaller than d_{eff} suggests that the GMR versus reduced magnetization M/M_S dependence should always be perfectly parabolic [5], i.e. the Co particles should be independent. This is the case, as indicated in one example in Fig. 4 (bottom), where the full line follows a parabolic law and perfectly reproduces the experimental data. This is true also for the sample annealed at $I = 5.5$ A, even if it exhibits a beginning of particles aggregation through magnetic (dipolar) interactions that give rise to larger effective clusters (see Figs. 2 and 3). This behaviour is confirmed by Fig. 4 (top) where the dipolar interaction energy of Co particles displays a maximum for the sample annealed at $I = 5.5$ A, but is always very far from room temperature thermal energy (dashed line), thus justifying the good SP behaviour of this alloy and its parabolic GMR versus reduced magnetization. The dipolar energy reduction could act to partially destroy nanoparticle clusters in agreement with the results of Fig. 2.

4. Conclusions

A full set of room temperature magnetic and magnetotransport measurements coherently describes the rapidly solidified $Cu_{95}Co_5$ alloy as superparamagnetic, with parabolic GMR versus reduced magnetization curves. However, detailed study of the Co particle effective size and distance shows that current annealing is able to strongly vary the number and size of Co particles, thus largely affecting the GMR value of the samples. Smaller, closer and non interacting Co particles situated at an average distance close to the conduction electron mean free path are responsible of the largest GMR ratio, while the strongest annealing conditions favour the redissolution of Co into the Cu matrix.

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