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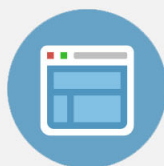
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Giant magnetoresistance and remanence in granular CoCu codeposited films

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We present experimental magnetoresistance, magnetization, and remanence data for CoCu granular films obtained by electron beam codeposition, subsequently submitted to a thermal treatment. The magnetization measurements show coexistence of superparamagnetic and ferromagnetic (or blocked superparamagnetic) Co grains. By modeling the experimental magnetization curves, the distribution function of the superparamagnetic particle size, the volume fraction of the blocked part of the Co particles, and their saturation magnetization are obtained. δM plots are constructed using the remanence curves. A correlation between the changes of the δM parameters and those of the giant magnetoresistance, caused by the annealing, is discussed. © 1998 American Institute of Physics. [S0021-8979(98)46811-7]

I. INTRODUCTION

The so-called granular ferromagnetics are composites of two materials, one of which consists of nanosized ferromagnetic particles (e.g., Fe, Co) dispersed in nonmagnetic metallic matrix (e.g., Ag, Cu, Au). Since the discovery of the effect of giant magnetoresistance (GMR) in such systems,^{1,2} they have been widely investigated due to their potential for practical applications and because they can be easily produced (usually sputtering, coevaporation, melt spinning, mechanical alloying, or electrodeposition), and their microstructure can be modified by suitable thermal treatment. The GMR in these granular systems is comparable to the ones found in multilayers. Extensive studies have been made to link the GMR response to the structural properties (e.g., interface, particle shape, size and density, intergranular distances, etc.).^{3,4}

The GMR effect in these systems is believed to be a manifestation of the conduction electron spin-dependent scattering on the local magnetic configuration. Qualitatively, it is assumed that the scattering probability is proportional to the correlation degree of the moments of adjacent pair of granules $\langle \mu_i \cdot \mu_j \rangle$.^{5,6} The resistance of the heterogeneous alloys is maximum in the case of random distribution, i.e., low correlation of magnetic moments at zero field. The applied field aligns the magnetic moments (high correlation) thus decreasing the resistance of the system.³

The magnetoresistive behavior of an homogeneous superparamagnetic (SPM) system has a relatively simple interpretation based on a description by the Langevin function. In this case it is given by $[L(\mu H/k_B T)]^2$, where H is the applied field, k_B the Boltzman's constant, and T the temperature. All the theories that use this description predict a dependence of the fractional magnetoresistance on the square of the reduced magnetization. However, this relation fails in some granular systems.⁷ Interparticle interactions⁸ and de-

pendence of the scattering process on cluster size⁹⁻¹¹ are the mechanisms that have been introduced to explain this discrepancy.

The effect of interactions on GMR of granular solids has been investigated,¹² using a model that obtain the moment configuration in the presence of dipolar magnetic fields. The GMR effect is found to be lowered by these interactions, and connected to low-range correlation of the magnetic moments. In this case, a deviation from the parabolic law is also observed. In the present work, the influence of the thermal treatment on the microstructure, magnetoresistance, and magnetic properties of CoCu codeposited films is investigated. The latter, along with the fact that the GMR for our samples do not saturate for the maximum magnetic field available, do not allowed us to use the deviations from the parabolic law to estimate the interaction effects. We used the technique based on the δM^{dc} plot, which has been extensively used recently, because it proved to be very sensitive to small changes in the remanence produced by interaction between the magnetic regions of the system.

II. EXPERIMENT

The 5000-Å-thick Co-Cu films with different compositions were codeposited by means of electron beam gun on Si (111) substrate. The nominal compositions are 10%, 20%, and 30% of cobalt. Thermal treatment was performed in a rapid annealing furnace under Ar atmosphere between 100 and 350 °C for 1 h. The magnetization and remanence curves were measured at room temperature using an alternating gradient magnetometer with the field applied parallel to the sample's plane. The magnetoresistance curves were measured in fields up to 6 kOe using the standard four points method.

III. RESULTS AND DISCUSSION

The conventional x-ray diffraction shows a pure fcc Cu structure, no traces of hcp Co are observed. Due to the small

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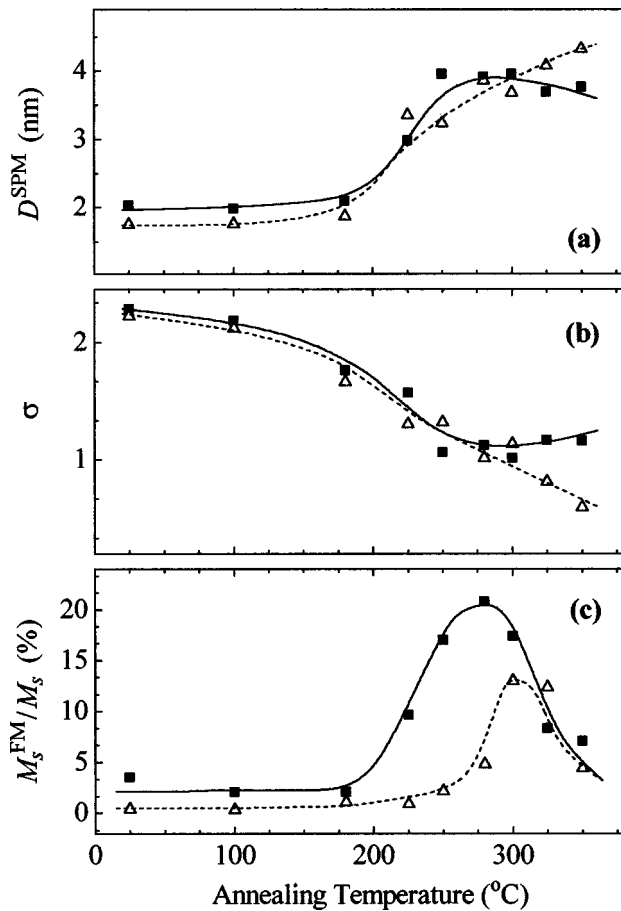


FIG. 1. (a) D^{SPM} and (b) σ vs. T_{ann} obtained from the SPM fit of the experimental magnetization curves for the samples with 10% and 20% Co concentrations; (c) The ratio between FM component and total magnetization; squares: 10% Co, and triangles: 20% Co. The lines are guide to the eye.

size of the Co particles and the high lattice coherency between the fcc Co phase and the Cu matrix, this Co phase cannot be distinguished.

The experimental magnetization curves show hysteresis. Here, as in our previous work,¹³ we consider the samples as consisting of two magnetic Co phases: (i) noninteracting SPM particles and (ii) “blocked” [interacting and/or larger ferromagnetics (FM)] grains. The sample magnetization can be written as $M(H) = M^{\text{FM}}(H) + M^{\text{SPM}}(H)$. The M^{SPM} term is a weighted superposition of Langevin functions assuming the following magnetic moment distribution function¹⁴

$$f(\mu) = \frac{N}{\sqrt{2\pi}} \frac{1}{\mu} \exp\left[-\frac{\ln^2(\mu/\mu_0)}{2\sigma^2}\right], \quad (1)$$

where N is the volumetric SPM particle density, μ_0 the particle magnetic moment, and σ the width of the distribution which differs from the log-normal one by the factor $1/\mu$. The mean value of μ is defined as $\langle\mu\rangle = \mu_0 \exp(\sigma^2/2)$.

In Figs. 1(a) and 1(b) the values of the mean SPM particle size, D^{SPM} , and σ used to fit the SPM components of the experimental magnetization curves are displayed as a function of the annealing temperatures, T_{ann} for the samples with 10% and 20% Co concentrations. Due to the low SPM

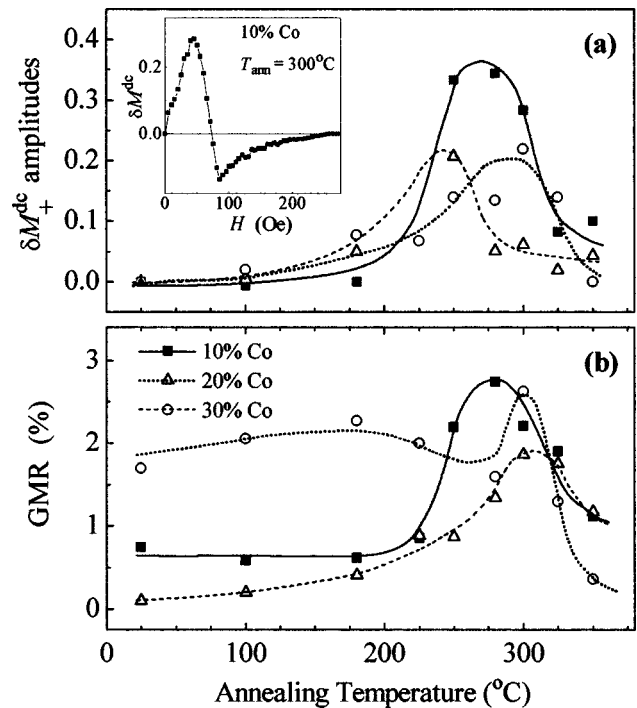


FIG. 2. (a) δM_+^{dc} vs. T_{ann} constructed from the remanent magnetization curves; the inset in this figure is a representative δM^{dc} plot for the sample with 10% Co, annealed at 300 °C. (b) GMR vs. T_{ann} for samples with different Co concentration. The lines are guide to the eye.

contribution to the total magnetization of the 30% Co sample, it was not possible to fit its SPM component. The dependence of the FM fraction, defined as M_s^{FM}/M_s , on T_{ann} is shown in Fig. 1(c) (the method of estimation of M_s^{FM} is described elsewhere).¹³

When the particles are sufficiently close to each other, the magnetic interactions can induce correlation between the adjacent moments, which may be both Ruderman–Kittel–Kasuya–Yosida (RKKY) and/or dipolar in nature. We tried to estimate this effect by using the maxima of the δM^{dc} plots, defined in our recent works.^{13,15} The positive amplitude values, δM_+^{dc} vs T_{ann} are represented in Fig. 2(a) (the negative ones almost do not change as a function of T_{ann} having values of ≈ 0.2). The inset in Fig. 2(a) shows a representative δM^{dc} plot (10% Co, $T_{\text{ann}} = 300$ °C). The reduced GMR values, defined as $[R(H) - R(H=0)]/R(H=0)$, obtained for $H_{\text{max}} = 6$ kOe, are shown in Fig. 2(b).

$T_{\text{ann}} \leq 200$ °C does not change significantly the morphology, magnetic, and magnetotransport properties of the samples. Figure 1(c) shows that in this T_{ann} range, the samples contain mostly SPM particles, with $D^{\text{SPM}} \approx 2$ nm. M_s values are rather smaller than M_s^{Co} for bulk Co, indicating that there is a large number of Co atoms dissolved in the Cu matrix.

Annealing at higher temperatures up to approximately 300 °C, results in a gradual increase of D^{SPM} and a decrease of σ , i.e., the SPM particles become larger and more uniform in size. Their number increases as well, as the total M_s values increase for all samples at the expense of the atomically diluted Co in the Cu matrix. GMR is enhanced, reaching its maximum for $T_{\text{ann}} \approx 300$ °C for all samples (for H_{max}

=6 kOe). This increase could be explained, assuming that the main contribution to the GMR comes from the random orientation of the SPM Co magnetic moments:² annealing at higher temperatures increases the number of SPM particles, changes their size, and the mean distance between them. The latter would become comparable to the mean free path of the conduction electrons, and more selective scattering would occur, thus increasing GMR. Although the FM fraction increases with increasing T_{ann} , it remains relatively small and does not influence the GMR, as observed from the magnetoresistance curves, which do not show rapid response at low fields and do not saturate at 6 kOe.

GMR of the samples annealed at $T_{\text{ann}} > 300$ °C decreases, despite that the D^{SPM} and σ do not change significantly. However, a decrease of the total M_s is observed for all samples, which can be explained by the decrease of the number of the SPM particles, as well as the increase of the distance between them, so the scattering event becomes infrequent and far apart, thus GMR is small. The decrease of M_s is not very clear, but we suggest that some Co is redissolved in the Cu matrix,¹⁶ which is expected to be present at high temperature.

All δM^{dc} plots are characterized by positive peaks in low fields and a relatively shallow minima in higher fields. There are two cases to be considered.

(i) the FM particles are mainly fcc Co, and weak positive (ferromagnetic, magnetizing) interactions present. This suggestion is supported by the similarity of the shape of the experimental δM^{dc} plots to that calculated for a disordered system of noninteracting cubic anisotropy particles with four easy magnetization axes,¹³ as well as by the statement of Kimoto *et al.*¹⁷ and Anno,¹⁸ that small Co particles have a fcc structure. In this case, the increase of δM^{dc} by increasing T_{ann} means that the interactions favoring parallel alignment weaken, and δM^{dc} tends to reach the value of ≈ 0.5 of that calculated for the noninteracting case plot.¹³ Thus, the maximum of the GMR, which is in the same T_{ann} range where δM^{dc} is maximum, can be associated with the weaker interactions, even that the FM fraction is not very large, having its maximum of $\approx 20\%$ for 10% and 20% Co.

(ii) The samples consist of hcp (uniaxial) Co. In such cases, positive deviations from the noninteracting zero line¹⁹ are attributed to strong FM interactions between the adjacent grains. The increase of δM^{dc} by increasing T_{ann} suggests increase of these interactions, and as result, GMR should be depressed, on the contrary of what is observed here. Therefore, we believe that the data support the first suggestion—the Co is mainly in fcc phase.

The reduction of δM^{dc} for the higher T_{ann} can be attributed not to an increase of the negative interactions, but mainly to an extinction of the FM phase, as indicated in Fig. 1(c).

IV. CONCLUSION

Experimental magnetoresistance and magnetization data for thermally treated CoCu codeposited films were reported. It was obtained that the GMR has maximum of $\approx 3\%$ (for $H_{\text{max}} = 6$ kOe) when the SPM particles have a specific size range and their size distribution width is relatively narrow. Assuming that the Co particles are mainly fcc, a correlation between the changes of the δM parameters and of GMR is observed. However, as the relative FM fraction is rather low in our samples, one should not assume that the magnetic interactions are the main, determining factor for the GMR changes.

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