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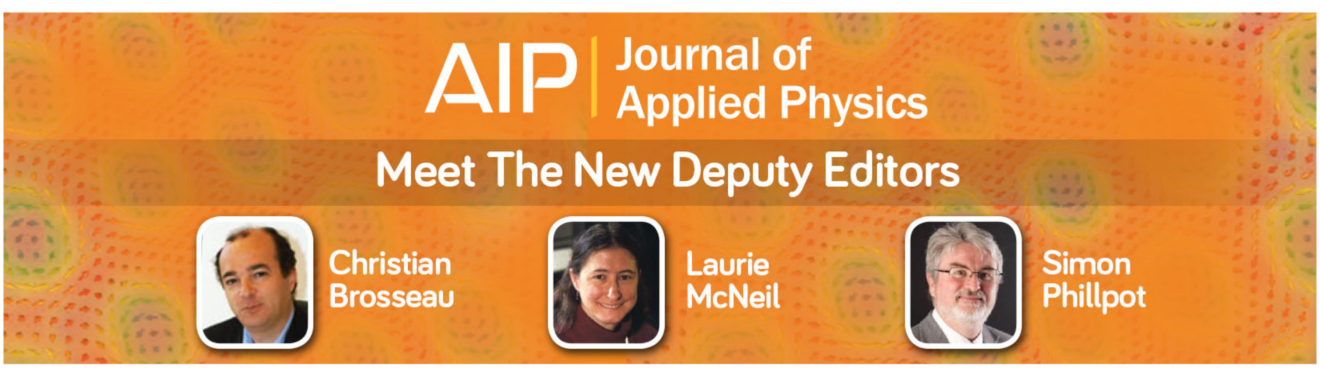

Short-time dynamics of correlated magnetic moments in superparamagnetic Cu–Co melt spun alloys exhibiting giant magnetoresistance

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Evidence for correlation among superparamagnetic particles in melt-spun $Cu_{100-x}Co_x$ systems ($x = 5-20$) exhibiting a giant magnetoresistance is obtained by plotting this quantity as a function of reduced magnetization. Two ranges, $R_\theta(H_e)$ and $R_\theta(H_e)$, have been recently introduced to describe the extent of correlation among angles of tilt (θ) and of twist (ϕ) of superparamagnetic moments precessing around a local field axis. The angle of tilt appears to be spatially correlated over a distance larger by a factor of 3 than the angle of twist. This difference is explained by analyzing the short-time dynamics of magnetic moments in superparamagnetic granular systems with long-range interactions (of dipolar and the RKKY-like type). The typical time constants characterizing the process of scattering of conduction electrons by adjacent magnetic moments (electronic time of flight, relaxation times for θ and φ) are discussed in detail. An explicit expression for $R_\varphi(H)$ is obtained by considering the competition between a magnetic interaction favoring parallel (or antiparallel) alignment or adjacent moments, and thermal disturbances resulting in a continuous loss of the phase coherence. [S0021-8979(97)69808-4]

Granular magnetic systems, where nanometer-sized, superparamagnetic particles of a ferromagnetic metal are dispersed in a nonmagnetic metal, developed in view of their prospective applications in magnetic recording, are still attracting considerable interest from the fundamentalist's viewpoint, owing to their isotropic giant magnetoresistance (GMR), often exhibited even at room temperature.[1] Among these materials, the $Cu_{100-x}Co_x$ system plays a relevant role, because it can be easily obtained in ribbon form over a wide composition range ($0<x\leq 20$) by planar flow quenching on a rotating drum, followed by proper annealing.[2,3] Varying the annealing conditions and/or methods[2,3] brings about a variety of granular structures characterized by rather different average values of the particle sizes. On the other hand, the width of the particle-size distribution appears to be affected to a lesser extent by annealing.[3] These results, indirectly obtained from room-temperature magnetization curves,[3] have been recently confirmed by independent low-temperature magnetic-susceptibility measurements.[4]

Plots of GMR versus reduced magnetization $m = M/M_s$ provide compelling evidence for magnetic correlation among superparamagnetic particles. In fact, the so-called reduced GMR, $(\Delta R/R)_{red}$ (a quantity defined in Ref. 3 and obtained by properly rescaling the GMR data) always displays a definite nonparabolic behavior when plotted against m, in disagreement with the prediction for uncorrelated magnetic particles, $(\Delta R/R)_{red} = 1 - m^2$. The typical flattening of actual $(\Delta R/R)_{red}$ vs m curves with respect to a parabola (see Fig. 1) has been explained by a phenomenological theory,[3] providing an expression for $(\Delta R/R)_{red}$ in systems characterized by superparamagnetic particles of magnetic moment μ:

$$\left(\frac{\Delta R}{R}\right)_{red} = 1 - \left\{ \overline{u^2} + (\langle u^2\rangle - \overline{u^2}) e^{-\lambda/R_\theta(H_e)} + \left[1 - \overline{u^2} - \frac{\langle u^2\rangle - \overline{u^2}}{1-\overline{u^2}} (1 - \overline{u^2} e^{-\lambda/R_\theta(H_e)}) \right] e^{-\lambda/R_\phi(H_e)} \right\}, \quad (1)$$

where $u = \cos\theta$, $\overline{u} \equiv m = L(x)$, $L(x)$ is the Langevin's function, and $x = \mu H_e/k_BT$, $\langle u^2\rangle = 1 - 2L(x)/x$, λ is an (effec-

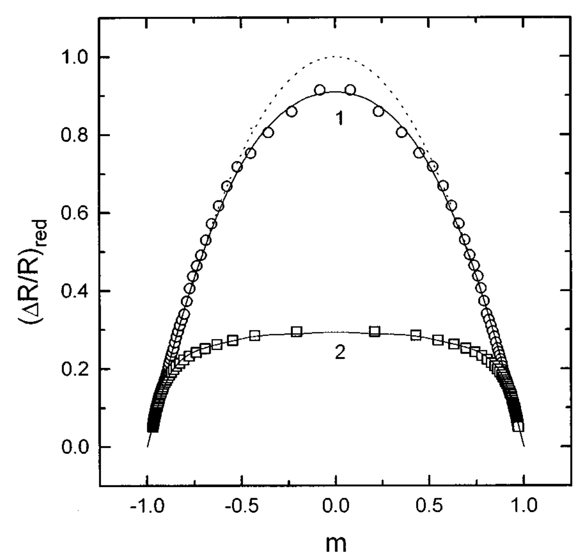

FIG. 1. Reduced GMR as a function of reduced magnetization m for $Cu_{90}Co_{10}$ (joule-heated, electrical current density $j = 4\times 10^7$ A/m^2 for 1 s; circles) and for $Cu_{80}Co_{20}$ (joule-heated, $j = 2\times 10^7$ A/m^2 for 1800 s; squares). Full lines: best fit according to Eq. (1). Dotted line: parabolic behavior.

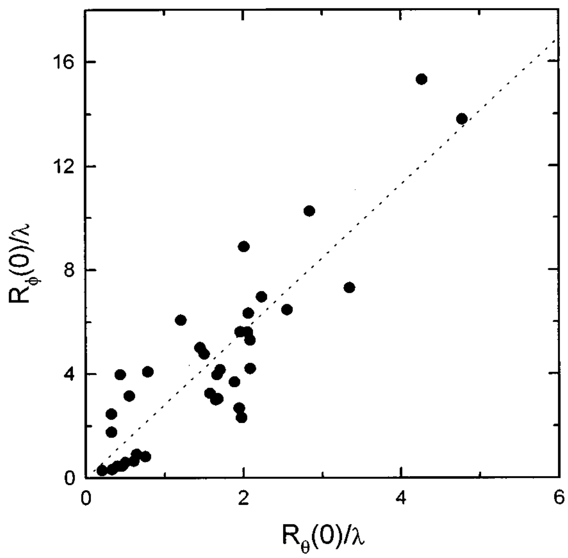

FIG. 2. Tilt-angle correlation length at zero external field, $R_\theta(0)$ as a function of twist-angle correlation length at zero field, $R_\phi(0)$, for 34 alloys of the $Cu_{100-x}Co_x$ system ($5 \leqslant x \leqslant 20$) submitted to various annealing treatments.

tive) electronic mean free path.[5] A central role is played by the quantities $R_\theta(H_e)$ and $R_\phi(H_e)$, i.e., the correlation lengths of tilt-angle and twist-angle fluctuations in the motion of magnetic moments around the local field axis, respectively. In spite of the success obtained applying Eq. (1) to fit actual $(\Delta R/R)_{\rm red}$ vs m curves (see Fig. 1), some aspects of this matter deserve further investigation.

First, the theory does not provide any explicit expression for $R_\phi(H_e)$, while $R_\theta(H_e)$ is given by[3]

$$R_\theta(H) = \sqrt{\frac{\mu^2}{k_B T}\,\alpha L'(x)}, \tag{2}$$

where $L'(x)$ is the first derivative of $L(x)$ with respect to its argument, and $\alpha = (1/3)\Sigma\lambda_{ik}\, r_{ik}{}^2$ contains the strength λ_{ik} of the coupling between particles i and k, where r_{ik} is the interparticle distance. For the sake of simplicity, $R_\phi(H_e)$ was given in Ref. 3 the same functional dependence on H_e as $R_\theta(H_e)$, but without any attempt of justifying this assumption.

Second, collecting the $R_\theta(0)/\lambda$ and $R_\phi(0)/\lambda$ values obtained from best fit of a number of experimental GMR vs m curves, and plotting one correlation length against the other, a linear relationship is clearly found, although the data are slightly scattered [see Fig. 2, where the plotted data refer to 34 alloys of the system $Cu_{100-x}Co_x$ $(5 \leqslant x \leqslant 20)$ and submitted to various annealing treatments[3]]. Note that the data reported here are comprehensive of the ones plotted in Fig. 16 of Ref. 3, where both scales were multiplied by a factor of $\sqrt{3}$ with respect to the present plot; this discrepancy is, however, irrelevant to the following discussion. A ratio $R_\theta(0)/R_\phi(0) \cong 3$ emerges from Fig. 2. Such a result has not been explained thus far.

The inadequacy of the model of independent moments can be related to two orders of reasons: First, the magnetic moments cannot be assumed as completely uncorrelated, owing to their long-range magnetic interactions; second, the time scale of the sequential scattering of a conduction electron by two adjacent magnetic moments is much shorter than the characteristic relaxation times associated to the changes in the moments' directions by effect of random disturbances, so that the electrons are actually sensitive to the instantaneous angle between the considered moments. As a matter of fact, the time of flight of a conduction electron between adjacent scatterers may be estimated to be of the order of 6×10^{-15} s with a Fermi velocity of about 10^8 cm/s and an average intercluster distance of about 6×10^{-7} cm, as experimentally observed in the $Cu_{100-x}Co_x$ system[3] (this is actually the distance between the borders of adjacent clusters; the average distance between centres is of the order of 1×10^{-6} m). On the other hand, the moments' directions in the superparamagnetic regime fluctuate in space by effect of both thermal and magnetic torques. Changes in the angle of tilt, involving changes in the local Zeeman energy, are related to the interaction between the localized moment and the thermal excitations of the lattice. Applying Néel's or Brown's models for the superparamagnetic relaxation,[6] and using the values $K \approx 1\times10^6$ erg/cm^3 for the magnetic anisotropy energy, $V \approx 7\times10^{-20}$ cm^3 for the average cluster volume, and a pre-exponential factor $f_0 \approx 10^{10}$ s^{-1}, a relaxation time $\tau_1 = f_0{}^{-1}\exp(KV/k_BT)$ of the order of 5×10^{-10} s is found at room temperature. The angle of twist is more sensitive to the fluctuations in the local field acting on each localized moment, as in the case of paramagnetic resonance and relaxation. The relaxation time for the angle of twist, τ_2, is of the order of $(\gamma H_i)^{-1}$, where γ is the gyromagnetic ratio and H_i is the rms magnetic field present on each site, arising from long-range interactions among moments, such as dipolar or RKKY-like interactions.[7] Using $H_i \approx 5\times10^2$ Oe (a value to be justified later), one finds $\tau_2 \approx 2\times10^{-10}$ s, not much smaller than τ_1. These values should be intended as merely representative quantities. The spread in the cluster sizes may bring about significant local deviations from these average values.

An expression for $R_\phi(H_e)$ can be obtained considering the short-time dynamics of magnetic moments. The main points of the theory follow: Let us consider a pair of magnetic moments (μ_i, μ_k), initially rotating with the same phase angle around the local field axis (supposed to be the same for both moments). We admit the existence of a weak interaction of the type $\epsilon_{ik} = -\lambda_{ik}\mu^2\cos\theta_{ik}$, favoring parallel (or antiparallel) alignment of moments, where θ_{ik} is the instantaneous angle between μ_i and μ_k and $|\lambda_{ik}| \approx 1/r_{ik}{}^3$ for both dipolar and RKKY-like interactions. If a change η_k in angle ϕ_k suddenly occurs at $t=0$, angle ϕ_i will relax toward the new ϕ_k in order to reduce the coupling energy. However, the corresponding relaxation rate, $\tau_{\rm rel} = \gamma\lambda_{ik}\mu\sin\theta_i\sin\theta_k$, where θ_i, θ_k are the tilt angles, is generally much smaller than $\tau_2{}^{-1}$, i.e., the rate at which the twist-angle coherence is lost. As a consequence, the relaxation process is continuously interrupted by new fluctuation events. Similar results are found considering the effect of z neighboring moments on the ith moment; a linear relation can be easily obtained between the twist-angle fluctuation η_i and the ones occurring at adjacent sites:

$$\eta_i = \gamma\mu\langle\sin^2\,\theta\rangle\tau_2'\sum_k\,\lambda_{ik}\eta_k, \tag{3}$$

where $\langle\cdots\rangle$ means the expectation value, $(\tau_2')^{-1}$ is the rate of loss of phase coherence between moment i and any of the z surrounding moments: $\tau_2' \cong \tau_2/z$, τ_2 is usually referred to a single pair of adjacent moments. In the presence of an external field, $\tau_2 \cong (\gamma H_t)^{-1}$, with $\mathbf{H}_t = \mathbf{H}_i + \mathbf{H}_e$. As a consequence,

$$\eta_i \cong \frac{1}{zH_t}\,\mu\langle\sin^2\,\theta\rangle\sum_k\,\lambda_{ik}\eta_k \tag{4}$$

at any time. Such a linear relation connecting fluctuations at neighboring sites allows one to write an expression for the correlation length $R_\phi(H_e)$:

$$R_\phi(H_e) \approx \left[\frac{1}{z}\,\frac{\mu}{H_i}\,\frac{2L(x)}{x}\,\alpha\right]^{1/2}, \tag{5}$$

where $\langle\sin^2\,\theta\rangle$ has been rewritten as $2\,L(x)/x$. When $H_e=0$, $R_\theta(0)$ and $R\phi(0)$ take the form

$$R_\phi(0) \approx \left[\frac{1}{z}\,\frac{\mu}{H_t}\,\frac{2}{3}\,\alpha\right]^{1/2}, \quad R_\theta(0) \approx \left[\frac{\mu^2}{k_BT}\,\frac{1}{3}\,\alpha\right]^{1/2}, \tag{6}$$

so that the ratio $R_\theta(0)/R_\phi(0)$ is

$$\frac{R_\theta(0)}{R_\phi(0)} = \left(\frac{\mu H_i}{k_BT}\,\frac{z}{2}\right)^{1/2}. \tag{7}$$

Note that this ratio does not contain the parameter α. The internal field H_i may be estimated (both for dipolar and RKKY-like interactions) as $H_i \cong (2\mu^2\Sigma_k 1/r_{ik}{}^6)^{1/2} \cong 2^{3/2}\mu/a^3$, where a is the average interparticle distance.[8] Typical values of H_i for the $Co_{100-x}Cu_x$ system are in the range $2\times10^2 - 1\times10^3$ Oe, depending on μ and a values. Taking values appropriate to the whole family of alloys studied ($\mu \approx 10^{-16}$ emu, $H_i \approx 3\times10^2$ Oe, $k_BT \approx 4\ 10^{-14}$ erg/atom, $z \approx 25$), one gets $R_\theta(0)/R_\phi(0) \approx 3$, in excellent agreement with the experimental result. The value $z \approx 25$ for the total number of magnetic particles effective in influencing the motion of the ith moment has been derived by comparing the values of all the parameters $R_\theta(0)$ [obtained by fitting the $(\Delta R/R)_{\mathrm{red}}$ vs m curves to Eq. (1)] with the corresponding theoretical expression, which can be rewritten as $R_\theta(0) \cong z^{1/2}[\mu^2/k_BT1/(3a)]^{1/2} \equiv z^{1/2}S$, because $\alpha \approx z/a$. Using the values for μ and a obtained from the fits of magnetization curves,[3,4] the quantity S is known for each alloy. Plotting $R_\theta(0)$ as a function of S, a linear relation of the type $R_\theta(0) \cong 5\ S$ is actually found.

Finally, two comments about the new expression for $R_\phi(H_e)$ are needed. First, substituting Eq. (5) to the expression used in Ref. 3 [formally identical to the one for $R_\theta(H_e)$] has only minor effects on the fits of experimental curves to Eq. (1). In particular, the values of $R_\theta(0)$ and $R_\phi(0)$ derived using Eq. (1) with the new functional dependence of $R_\phi(H_e)$ always remain almost coincident with the ones obtained in Ref. 3. The second comment is about the different characters of the expressions for the two angular correlation ranges [Eqs. (2) and (5)]. In fact, twist-angle correlation occurs between instantaneous ϕ values, while tilt-angle correlation is between expectation values of $u \equiv \cos\theta$, as evidenced by the presence of the $L'(x)$ function. Such a difference reflects the fact that fluctuations in θ and in ϕ occur via quite different mechanisms. Thermal excitations are needed to induce changes in θ, involving an exchange of energy between a magnetic moment and the environment. The effects of such excitations on adjacent moments are completely random, so that instantaneous tilt-angle fluctuations are essentially uncorrelated. A spatial correlation exists, therefore, only between expectation values of u. On the contrary, the local field $\mathbf{H_i}(\mathbf{r},t)$, whose fast changes with time are responsible for twist-angle fluctuations, is weakly dependent on position over distances of the order of the interparticle distance, so that instantaneous fluctuations of ϕ on adjacent moments may be substantially correlated, as shown in this paper.

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