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Giant magnetoresistance for superparamagnetic particles: Melt-spun granular CuCo

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We have measured the giant magnetoresistance (GMR) and the magnetization of a melt-spun granular sample of $Cu_{87}Co_{13}$. The Cu matrix contains very small particles of Co that exhibit superparamagnetism. Although the GMR is due to these small superparamagnetic particles, we find that the GMR does *not* vary quadratically with the magnetization. This unexpected result is attributed to the presence of a range of sizes for the superparamagnetic particles. Assuming a simple distribution of particle sizes, we calculated the magnetic-field dependence of the GMR and find excellent agreement with experiment.

An interesting approach to the study of the giant magnetoresistance (GMR) was initiated in 1992, when it was demonstrated[1,2] that a GMR is exhibited by a nonmultilayer heterogeneous sample containing ferromagnetic granules embedded in a nonmagnetic matrix. Since these pioneering studies, the GMR of such heterogeneous systems has been a subject of great interest.[3–12] If the ferromagnetic particles are sufficiently small, then they become superparamagnetic (SPM).[13] Gittleman *et al.*[14] showed many years ago that SPM particles exhibit several distinctive magnetoresistance properties. In particular, these workers concluded that the magnetoresistance should be proportional to the *square* of the magnetization. As a result, for granular systems, the GMR data are often plotted as a function of the magnetization in the expectation that parabolic (quadratic) behavior will be observed. In this paper, we show that such a quadratic dependence is *not* necessarily obtained. We have prepared a melt-spun granular sample of CuCo and measured its GMR and its magnetization as a function of magnetic field. We find that the GMR has nearly a *linear* dependence on the magnetic field at high temperatures, and there is *no* temperature regime in which the field dependence of the GMR is proportional to the field dependence of the square of the magnetization.

This surprising result is attributed to the presence of a range of sizes for the very small ferromagnetic particles, with a portion of the particles being "blocked" (and hence not SPM). As the temperature is increased, a progressively larger fraction of the ferromagnetic particles becomes "unblocked" (and hence SPM), but some particles remain blocked even at room temperature. Assuming a simple distribution of particle sizes, we calculated the magnetic-field dependence of the GMR and find excellent agreement with experiment.

Ingots of the alloy $Cu_{87}Co_{13}$ were produced by arc melting Co (99.99%) and Cu (99.999%) in an argon-arc furnace. The rapid quenching of the sample was achieved by depositing the molten alloy onto a rapidly rotating copper wheel in 0.5 atm of helium gas. This procedure produces ribbons that are several cm in length, 1–2 mm in width, and about 30 μm thick. Stoichiometry and uniformity of composition were routinely monitored by electron microprobe and were found to vary by less than 0.5 at. %.

In Fig. 1, we display our measured values for the GMR as a function of the magnetic field for a series of temperatures. The normalized magnetoresistance $\delta R/R$ is given by the usual definition,

$$\delta R/R = \frac{R(B)-R(0)}{R(0)}, \tag{1}$$

where $R(B)$ is the resistance of the sample in magnetic field B. We note from Fig. 1 that above 100 K, the data for $\delta R/R$ are nearly linear out to several tesla, whereas for lower temperatures, no such linear regime is seen. Thus, at the higher temperatures, $\delta R/R$ is clearly *not* proportional to $[M(B)]^2$, the square of the magnetization. However, as we shall see, at the lower temperatures as well, $\delta R/R$ is not proportional to $[M(B)]^2$.

In Fig. 2, we compare the measured values of $\delta R/R$ with the calculated values at three representative temperatures. The agreement between theory and experiment is evident from the figure. The quality of the agreement is the same for all eight temperatures. Note in particular that the same calculation reproduces *both* the nearly linear behavior of $\delta R/R$ at high temperatures, as well as the clearly nonlinear behavior at the lower temperatures.

The key to understanding these results lies in the fact that the ferromagnetic Co particles are present in the Cu matrix in a range of sizes, and as the temperature increases, the Co

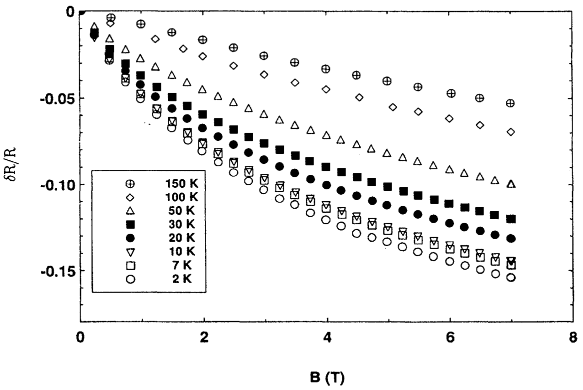

FIG. 1. Magnetic-field dependence of the GMR, measured at eight different temperatures.

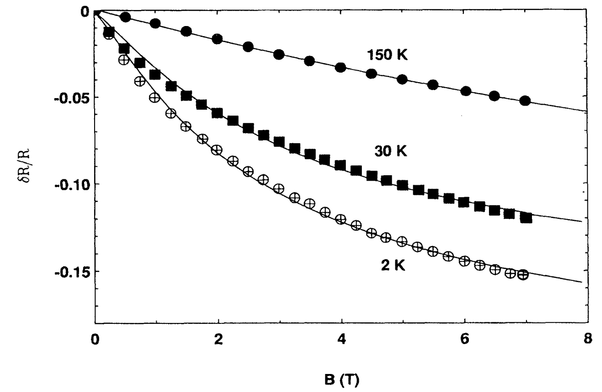

FIG. 2. Magnetic-field dependence of the GMR at three temperatures. The symbols represent the data and the curves give the calculated values.

particles become progressively unblocked and hence SPM. This is clearly seen, for example, in the temperature dependence of the magnetization data measured under conditions of field cooling and zero-field cooling. We find for our sample that the field-cooled data always lie above the zero-field-cooled data, even at room temperature. This, together with other aspects of the magnetization data, indicates that there exists a range of blocking temperatures—up to room temperature—with a corresponding range of particle sizes. A full discussion of the magnetization data is the subject of a separate publication.[15]

The GMR is generally believed to be due to spin-dependent electron scattering, as the electron moves from one Co particle to a neighboring one. The probability for this spin-dependent scattering depends on $\langle\cos\theta\rangle$, the thermal average value in field B of the angle θ between the magnetic moments of the initial and final Co particles traversed by the electron.[14] Since the value of $\langle\cos\theta\rangle$ for SPM particles is proportional to the square of $M(B)$, hence so is $\delta R/R$.

The above result applies only if the Co particles are SPM. However, if a range of particles sizes are present, then there will be a corresponding range of blocking temperatures. Therefore, at any given temperature, only a fraction of the Co particles will be SPM (unblocked), while the remainder will not be SPM (blocked). When considering electron scattering from one Co particle to another, one must thus distinguish between three cases: (i) both Co particles are SPM, (ii) neither Co particle is SPM, and (iii) one of the Co particles is SPM, while the other is not. Each of these three cases makes a completely different contribution to the field dependence of $\delta R/R$, for the following reason. For a SPM particle, a large magnetic field, of order many tesla, is needed to align its magnetic moment. By contrast, the moment of a blocked (non-SPM) particle is aligned in a very much smaller field.

Case (i) is the case that is usually considered; it makes the usual contribution to $\delta R/R$ and need not be discussed further. For case (ii), the moments of both Co particles are aligned at relatively small fields, and the magnetic field then has no further effect on the electron scattering or, equivalently, on the resistivity. Therefore, case (ii) makes *no* contribution at all to $\delta R/R$ at larger fields.

TABLE I. Values of the parameters at different temperatures.

Temperature (K)	$\mu_{\max}$ (Bohr magnetons)	α	β
2	7	0.106	0.00
7	20	0.136	0.00
10	27	0.146	0.00
20	47	0.151	0.00
30	64	0.143	0.00
50	86	0.125	0.00
100	112	0.096	0.01
150	126	0.082	0.02

The new feature of the present analysis is case (iii), which we find to make the dominant contribution to $\delta R/R$. This explains why the expected behavior for $\delta R/R$, based on case (i), is not observed. For case (iii), the moment of the non-SPM Co particle is quickly aligned, and thus at high fields, $\langle\cos\theta\rangle$ depends on the alignment of only *one* SPM particle. This implies that $\delta R/R$ contains a term that depends *linearly* on the magnetization, which is in accord with the data.

These ideas can be made quantitative. The complete analysis of $\delta R/R$ takes into account that there are, in fact, two independent electron currents present ("two-current" model), corresponding to spin-up electrons and spin-down electrons. Moreover, one must say something about the distribution of sizes for the Co particles. In the absence of specific information, we simply assumed that the contribution to the magnetization arising from the particles whose magnetic moment lies between μ and $\mu+d\mu$ is independent of μ, up to a maximum value of $\mu_{\max}$. This implies that as the particle size increases, their number decreases correspondingly to preserve the same moment for each increment $d\mu$.

The calculation of $\delta R/R$ is conveniently carried out within the framework of the "resistor network" model of Edwards, Mathon, and Muniz.[16,17] A tedious but straightforward calculation (details to be published separately[18]) yields the following result:

$$\delta R/R = -\alpha(J+I) - \beta(J^2+2I+3I^2), \tag{2}$$

where

$$J(B) = X^{-1}\ln(\sinh X/X), \tag{3}$$

$$I(B) = X^{-1}\int_0^X dx[\tfrac{1}{3} - x^{-1}L(x)], \tag{4}$$

$$X(B) = B\mu_{\max}(T)/k_B T. \tag{5}$$

Here, $L(x)$ is the Langevin function $(=\coth x - 1/x)$ and $\mu_{\max}(T)$ is the magnetic moment of the largest Co particle that is unblocked (SPM) at temperature T.

The values of $\mu_{\max}(T)$ (listed in Table I) increase with temperature because of the progressive unblocking of larger Co particles at higher temperatures. These values were previously determined[15] from the magnetization data, and hence they are *not* adjustable parameters in the calculation of $\delta R/R$. In Fig. 3, we illustrate the quality of the agreement be-

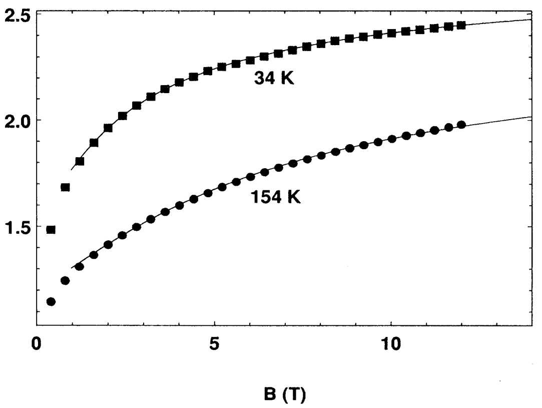

FIG. 3. Magnetic-field dependence of the magnetization at two temperatures. The symbols represent the data and the curves give the calculated values.

tween theory and experiment that we obtained for the magnetization $M(B)$ at two temperatures close to those used in Fig. 2.

Equations (2)–(5) constitute an explicit expression for the magnetic-field dependence of $\delta R/R$ in terms of two parameters, α and β, which give the magnitude of the contributions to $\delta R/R$ due to electron scattering for case (iii) and for case (i), respectively. The values of these two parameters were determined by a nonlinear least-squares fit to the $\delta R/R$ data and are listed in Table I. The resulting calculated values of $\delta R/R$ are given by the curves shown in Fig. 2.

It is seen from Table I that the value of β is negligible below 100 K and remains much smaller than α even at the highest measured temperature. This confirms our earlier assertion that case (iii) scattering makes the dominant contribution to $\delta R/R$ and the widely discussed case (i) scattering is, in fact, unimportant for our sample.

We note from Table I that α first increases with temperature and then decreases. This can be understood as follows. The initial increase in α is due to the increased probability of having SPM particles of Co as the temperature is increased. In other words, with increasing temperature, one moves from case (ii) scattering (*neither* Co particle is SPM) to case (iii) scattering (*one* of the Co particles is SPM). At very low temperatures, the chances are negligible of having case (i) scattering (*both* Co particles are SPM) and hence β is vanishingly small.

At higher temperatures, the value of α decreases for two reasons. First, as the temperature increases, one moves from case (iii) scattering to case (i) scattering, and second, there is a general tendency for $\delta R/R$ to decrease at higher temperatures because of the occurrence of additional spin-independent scattering events (such as electron-phonon scattering) as well as spin-flipping scattering events (such as electron-magnon scattering). Thus, the temperature dependence of both α and β conform to our expectations.

In summary, we have measured the GMR of a melt-spun granular sample of $Cu_{87}Co_{13}$ that contains very small superparamagnetic particles. A consistent picture emerges from the magnetic-field dependence of the GMR and that of the magnetization. In particular, the GMR tends to vary *linearly* as the magnetization. This behavior is due to the fact that the small Co particles are present in a wide range of sizes.

Assuming a simple distribution of particle sizes, we calculated the field dependence of the GMR of the superparamagnetic particles and find excellent agreement with experiment.

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