Giant Magnetoresistance in Nonmultilayer Magnetic Systems

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We have observed isotropic giant magnetoresistance (GMR) in nonmultilayer magnetic systems using granular magnetic solids. We show that GMR occurs in magnetically inhomogeneous media containing nonaligned ferromagnetic entities on a microscopic scale. The GMR is determined by the orientations of the magnetization axes, the density, and the size of the ferromagnetic entities.

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Recently a great deal of attention has been focused on giant magnetoresistance (GMR), first observed in Fe/Cr multilayers [1] and subsequently in many other multilayers (e.g., Co/Cu, Fe/Cu) [2-5] and sandwiches (e.g., Co/Au/Co) [6]. Equally intriguing is the presence of the oscillatory interlayer magnetic exchange coupling in many multilayers exhibiting GMR [1-3,7]. In the multilayers with antiferromagnetic (AF) alignment (in zero external magnetic field), the electrical resistance can be as much as 50% larger than that with the ferromagnetic alignment (achieved by applying a large field). Even though GMR has been observed in many multilayer systems, the fundamental mechanisms responsible for GMR remain elusive, and so far only phenomenological models have been proposed [8-10]. It is important to stress that, to date, *all* GMR's have been observed in multilayer geometry involving either transition-metal elements or alloys. Layer structure continues to be featured in all theoretical and experimental investigations of GMR. Some models of GMR have highlighted electronic effects in the form of oscillatory density of states or oscillatory exchange interactions [9]. A more widely discussed model of GMR is based on interfacial spin-dependent scattering [8,10]. The importance of the roughness of the interfaces in the layer structure has been stressed [5].

It should be emphasized that, in the layer geometry, the ferromagnetic layer is always contiguous, with large connecting ferromagnetic domains. In this work, we have chosen *nonmultilayer* magnetic media to investigate a number of critical and hitherto unexplored aspects concerning GMR. Specifically, we wish to explore the extent to which GMR depends on the layer structure, the connectivity of the ferromagnetic entities, the size of the magnetic domains, and the relationship between GMR and magnetic alignment. The key feature of our magnetic media is that they are materials consisting of immiscible elements from which both homogeneous metastable alloys and granular magnetic solids can be made. Consequently, we can explore the prospects of GMR in homogeneous ferromagnetic alloys as well as magnetic granular systems consisting of nonconnecting ferromagnetic entities of various sizes. Our results show that GMR can be readily observed in nonmultilayer but magnetically inhomogeneous media, in which single-domain ferromagnetic entities exist. The size of the ferromagnetic particles can be optimized in order to maximize GMR. We also show that the GMR is directly linked to the global magnetization of the specimen, and the directions of the magnetization axes of the ferromagnetic particles.

While Fe and Co form alloys with most other metallic elements, they are immiscible with a few, among them Cu. This is especially relevant since multilayers of Fe/Cu and Co/Cu have already exhibited GMR. Equilibrium phase diagrams of Fe-Cu and Co-Cu allow negligible solubility. However, homogeneous metastable alloys of Fe-Cu and Co-Cu over the entire composition range can be made by vapor quenching, i.e., vapor deposition using a low substrate temperature as previously described by some of us [11,12]. Annealing a metastable alloy at an elevated temperature (T_A) [or depositing films at an elevated substrate temperature (T_S)] causes the formation of a granular magnetic system consisting of single-domain ferromagnetic particles embedded in a metallic medium (Cu) [13,14]. The size of the ferromagnetic particles, ranging from a few to a few tens of nm, can be controlled by T_A or T_S.

We have used a dc magnetron sputtering device with a fixed substrate temperature (T_S) between 77 and 900 K. Each sample has been sputtered from a single composite target. A number of Co-Cu, Fe-Cu, and Gd-Ti samples have been deposited onto glass substrates. To avoid any possible thin-film effects, films between 4 and 10 μm thick have been made. The details of fabrication, x-ray diffraction analyses, and transmission electron microscopy have been published elsewhere [11-14]. Electrical resistance has been measured at temperatures between 5 and 300 K in a four-terminal geometry with an in-plane current. The magnetic field (up to 50 kG) has been applied parallel to the current ($\rho_{\parallel}$) and perpendicular to the film plane ($\rho_{\perp}$). The magnetization and hysteresis loop have been measured using a SQUID magnetometer. In the following we will describe the results of some representative samples. In this context, no GMR means it is less than 1%. We will confine the discussions mainly to the low-temperature data where effects due to scattering by phonons and magnons are small.

The magnetic phase diagram of metastable homogeneous Co_xCu_{100-x} alloys has recently been determined

[12]. Samples of Co_xCu_{100-x} with $x=16$, 20, 38, and 80 are used here. All the samples have been sputtered at $T_S=77$ K, plus $Co_{16}Cu_{84}$, which has also been sputtered at $T_S=350\,°C$. Those sputtered at $T_S=77$ K with $x=16$ and 20 are spin-glass-like below 20 K, and those with $x=38$ ($T_c \approx 400$ K) and $x=80$ (T_c in excess of 700 K) are ferromagnetic.

In the $Co_{16}Cu_{84}$ sample ($T_S=350\,°C$), phase separation occurs resulting in small fcc ferromagnetic single-domain Co particles embedded in the Cu medium. The GMR data are displayed as $\Delta\rho(H)/\rho=[\rho(H)-\rho(0)]/\rho(0)$, where $\rho(0)$ is the value in the initial $H=0$ state. GMR as large as 9% at 5 K has been observed as shown in Fig. 1(a), demonstrating that GMR is not inherent to the multilayer geometry. The GMR results are closely correlated with the measured hysteresis loop shown in Fig. 1(b). The corresponding points (a, b, c, d, and e) are labeled in both graphs, sharing the same horizontal (field) axis. A maximum in resistance is observed at the coercive field c and e where the magnetization (M) is zero, and the smallest resistance is observed at b and d, when a complete alignment of the magnetization vectors of all the Co particles is achieved. It is important to note that here M is the *global magnetization* of many separate Co particles. When $M=0$, all the moments within each single-domain Co particle remain ferromagnetically aligned, but the global magnetization, summed over all the particles, is zero.

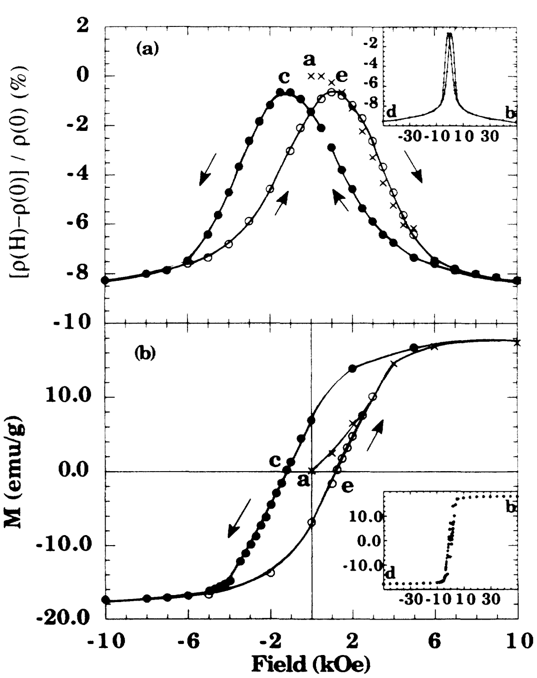

FIG. 1. Perpendicular magnetoresistance $[\rho(H)-\rho(0)]/\rho(0)$ as a function of external field (H) and hysteresis loop at 5 K for the phase-separated $Co_{16}Cu_{84}$ sample deposited at $T_S=350\,°C$. The crosses, open circles, and solid circles denote the initial curve ($a \rightarrow b$), and branches of decreasing ($b \rightarrow c \rightarrow d$) and increasing fields ($d \rightarrow e \rightarrow b$).

To explore the possible dependence of GMR on the size of the Co particles, samples of $Co_{20}Cu_{80}$ have been annealed for 10 min at successively higher temperatures of $T_A=200$, 500, and 650 °C, resulting in progressively larger particle sizes. The GMR of 11.5%, 16.5%, and 9%, respectively, have been observed as shown in Fig. 2. With increasing Co particle size, the GMR first *increases* and then *decreases*, illustrating a size range in which GMR is maximized. Also noted in Fig. 2, the saturation of GMR in the sample with $T_A=200\,°C$ is more sluggish, whereas in the sample with $T_A=600\,°C$ the saturation is more clear-cut.

With the phase-separated sample of $Co_{38}Cu_{62}$ annealed at $T_A=480\,°C$, a GMR of 13% has been observed at 5 K. Thus, in all the Co-Cu samples mentioned above, large GMR's have been observed in magnetically inhomogeneous media. In the homogeneous ferromagnetic alloy of $Co_{80}Cu_{20}$ ($T_S=77$ K), there is no GMR (magnetoresistance of $\approx 0.5\%$). We have also measured a phase-separated $Fe_{30}Cu_{70}$ ($T_A=350\,°C$) alloy to ascertain that the results observed in Co-Cu are not unique to that system. Indeed, the phase-separated sample of $Fe_{30}Cu_{70}$ shows a GMR of 9% at 5 K. However, in the phase-separated $Gd_{25}Ti_{75}$ sample, where Gd and Ti exist as separate hcp phases due to their immiscibility, *no* GMR has been observed. The GMR's of all the samples decrease with increasing temperatures. For example, the values of GMR of $Co_{38}Cu_{62}$ ($T_A=480\,°C$) at 5 and 300 K are 13% and 8%, respectively. The behavior of GMR, depending on composition, annealing conditions, and temperature, will be described elsewhere.

From the above results several conclusions emerge. First of all, GMR occurs in *magnetically inhomogeneous media containing nonaligned ferromagnetic entities on a microscopic length scale* (Λ), *roughly the mean free path*. This conclusion accounts for the lack of GMR in homogeneous ferromagnetic alloys, and the observation of

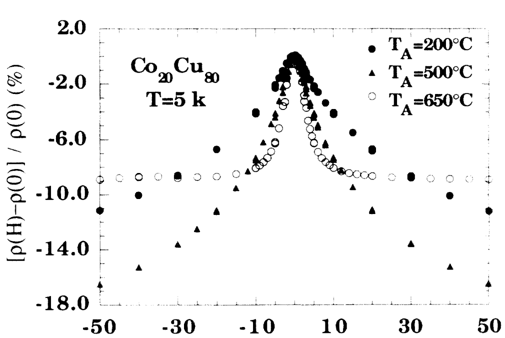

FIG. 2. Perpendicular magnetoresistance $[\rho(H)-\rho(0)]/\rho(0)$ at 5 K as a function of external field (H) for the phase-separated $Co_{20}Cu_{80}$ sample annealed at $T_A=200$, 500, and 650 °C for 10 min.

GMR in inhomogeneous magnetic granular media. In the metastable homogeneous $Co_{80}Cu_{20}$ alloy, the ferromagnetic domains are very large compared with Λ; consequently, no discernable GMR can be observed. GMR appears when the sample is magnetically inhomogeneous with nonaligned ferromagnetic entities, provided that the inhomogeneity occurs on a length scale comparable to Λ. In the granular systems with isolated ferromagnetic entities, high electrical resistance occurs when the magnetic axes of the particles are not aligned. Low resistance is obtained when the magnetic axes are aligned. The dependence of GMR on particle size can also be understood. When phase separation first occurs at a low annealing temperature, the Co particles are small and few in number. Scattering events within Λ are infrequent and far apart, thus GMR is small. The magnitude of GMR grows with increasing number and particle sizes. When the particle sizes become large compared with Λ, GMR is degraded.

We should emphasize the key features of magnetic granular solids. GMR in granular systems is *isotropic*. Both the perpendicular and longitudinal magnetoresistances are essentially the *same*, whereas in multilayers they are very different because of the demagnetizing factor [1]. In a granular system with small single-domain entities, the sizes of the ferromagnetic domains remain unchanged. The external field only rotates the magnetic axes of the particles. The rotation towards a complete alignment of all magnetic axes gradually reduces the resistance. The hysteresis loop of a granular magnetic system is precisely a signature of the rotation of the magnetic axes. The close correlation between the two branches of GMR and the hysteresis loop shown in Fig. 1 attests to this fact. To further illustrate this connection, we combine the two GMR branches and the hysteresis loop as GMR versus global magnetization as shown in Fig. 3. The size of the GMR is clearly linked to the global magnetization measured along the external field direction.

Since maxima in resistance occur at $H=\pm H_c$, we use $[\rho(H)-\rho(H_c)]/\rho(H_c)$ instead of $[\rho(H)-\rho(0)]/\rho(0)$. Note that the difference between $\rho(0)$ and $\rho(H_c)$ is very small. The GMR results can be approximately described as

$$[\rho(H)-\rho(H_c)]/\rho(H_c)\approx -A(M/M_s)^2, \qquad (1)$$

where M is the global magnetization and M_s the saturation magnetization. The prefactor A determines the ultimate size of the GMR. Its magnitude depends on spin-dependent scattering, as well as the number and the sizes of the ferromagnetic entities within Λ. The $(M/M_s)^2$ dependence is illustrated by the solid curve $[\Delta\rho/\rho=-0.065(M/M_s)^2]$ shown in Fig. 3. Apart from some deviations at $(M/M_s)^2\approx 1$, the agreement is very good. In a granular system, $(M/M_s)^2=\langle\cos\theta\rangle^2$, where θ is the angle between the magnetization axis of a particle and the external field, and $\langle\cos\theta\rangle$ is averaged over many ferromagnetic entities.

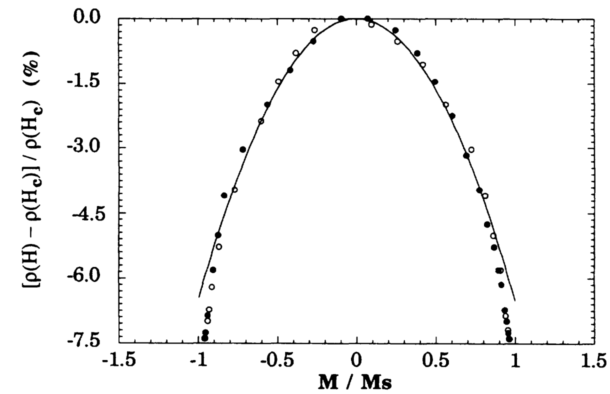

FIG. 3. Magnetoresistance $\Delta\rho/\rho=[\rho(H)-\rho(H_c)]/\rho(H_c)$ vs global magnetization at 5 K for the phase-separated $Co_{16}Cu_{84}$ sample taken from Fig. 1. Open and solid circles are those with increasing and decreasing fields, respectively. The solid curve is $\Delta\rho/\rho=-0.065(M/M_s)^2$ as mentioned in the text.

The GMR is the *extra* electrical resistance due to scattering from nonaligned ferromagnetic entities. It should be emphasized that the crucial factor for GMR is $\langle\cos\phi_{ij}\rangle$, where ϕ_{ij} is the angle *between* the axes of the ferromagnetic entities. It is simple to show that $\langle\cos\phi_{ij}\rangle=\langle\cos\theta\rangle^2$, when the moments of the particles are uncorrelated. This suggests that models for GMR in granular systems should include the *relative orientations* of the magnetic axes of the ferromagnetic entities. As suggested by Eq. (1), in a magnetically inhomogeneous medium, the resistivity at low temperatures varies as $\rho=\rho_0+\rho_m[1-(M/M_s)^2]$, where the first term ($\rho_0$) is the residual resistivity, and the second term depends on the global magnetization.

Here the factor $(M/M_s)^2$ is an approximation to illustrate the underlying effect. In this description, both AF alignment and randomly oriented magnetization axes would give the same GMR, since $M=0$ in both cases. However, experimental evidences suggest that a larger GMR is realized with more random axes. As shown in Fig. 1, the largest resistance of granular $Co_{16}Cu_{84}$ is not observed at the coercive fields (points c and e) with $M=0$, but at the original unmagnetized state (point a) also with $M=0$ and randomly orientated magnetic axes. This effect has also been seen, although seldom discussed, in multilayer systems where the largest resistance is obtained at the initial $H=0$ state before applying a magnetic field [4].

It is worth mentioning that Dieny *et al.* show in the exchange-biased layers that nonaligned layer structure *and* a sizable GMR can both be achieved without the intricate AF interlayer coupling [15]. Also noteworthy, they [15] and Chaiken, Prinz, and Krebs [16] have shown that the GMR is correlated with the *angle* of misalignment of the magnetization axes of the layers, which is consistent with the present results.

Finally, we comment on the null GMR results of granular Gd-Ti samples, in which ferromagnetic entities exist. Gd is a simple ferromagnet with localized S-state moments. Before additional rare earth systems are examined, which is underway, it would be premature to conclude that GMR is exclusive to $3d$ metal systems. Nevertheless, one notes the $3d$ moments have a strong itinerant nature, distinctively different from those of the rare earths. A lack of GMR in the $4f$ elements may not be totally surprising.

In summary, we have demonstrated that GMR is not unique to magnetic multilayers. Using immiscible alloy systems, GMR can be readily observed in magnetically inhomogeneous media with nonaligned ferromagnetic entities. The value of GMR depends on the density and the size of the ferromagnetic entities.

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