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Giant magnetoresistance in the granular Co-Ag system

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We report giant magnetoresistance (GMR) as much as 75% at 5 K and 24% at room temperature in granular Co-Ag. We show that the GMR is isotropic and is the consequence of the departure from ferromagnetic alignment of the Co particles. We have also determined the resistivity component responsible for GMR and its temperature dependence.

A recent intriguing discovery is giant magnetoresistance (GMR), first realized in Fe-Cr multilayers,[1] and subsequently in many other multilayers.[2-4] GMR presents a challenge for the fundamental understanding of scattering processes as well as having potential applications in magnetoresistive devices. Many experimental observations of GMR are in the multilayer geometry, upon which many theoretical models[5-7] are also based. Very recently, GMR has been observed by Berkowitz *et al.*[8] and Xiao *et al.*[9] in nonmultilayer but granular media consisting of small ferromagnetic single-domain Co particles in a medium of Cu. This development provides a new perspective in the understanding of the mechanisms for GMR.

In this paper we report results of GMR in a different granular magnetic system, namely, Co in an Ag medium, where GMR values as much as 75% have been observed. We show that the GMR is essentially isotropic, independent of the direction of the applied field with respect to that of the current. Equally important, we have separated out the resistivity component responsible for GMR from the total resistivity, and determined its temperature dependences.

The phase diagram of Co and Ag shows that they are mutually insoluble, no stable alloys of appreciable compositions exist under equilibrium conditions. However, it has been previously shown that metastable alloys of immiscible elements can be fabricated by vapor quenching.[10-13] Upon annealing the metastable alloys at elevated temperatures, phase separation occurs, causing the formation of small single-domain ferromagnetic particles embedded in a metallic medium, i.e., a granular magnetic solid.[11-13] The size and density of the Co particles are dictated by the annealing temperature and the volume fraction of Co. We have purposely chosen materials with a low Co volume fraction, so that the Co particles remain isolated in the Ag medium.

We used a dc magnetron sputtering system to fabricate metastable $Co_{20}Ag_{80}$ alloy samples (Co volume fraction =20%, Co atomic fraction =28%) onto substrates held at 77 K. Each sample, several μm in thickness, was subsequently annealed at various temperatures (T_A) in a high-vacuum furnace for 10 min. X-ray diffraction patterns of these samples are shown in Fig. 1. The as-prepared sample is a metastable fcc alloy. After annealing, a second set of fcc diffraction peaks emerges, indicating the formation of small fcc Co particles in fcc Ag. At successively higher T_A, the Co particles become larger, resulting in a narrower diffraction peak. These microscopic features are verified by transmission electron microscopy. Converging-beam electron diffraction and energy dispersive analyses both confirm that Co particles are surrounded by Ag.

Electrical resistivity [$\rho(H,T)$] has been measured as a function of magnetic field (H) and temperature (T) using a standard four-terminal configuration with an in-plane current. Measurements were done with the magnetic field (**H**) perpendicular to the film plane (perpendicular $\rho_\perp$), **H** parallel to the current (longitudinal $\rho_\parallel$), and **H** in-plane and perpendicular to the current (transverse ρ_T). The sample dimensions were determined using a profilometer. In Fig. 2 we show the magnetoresistance of these samples at 5 K. The data are presented in the fractional form of $\Delta\rho/\rho=[(\rho(H,T)-\rho(0,T)]/\rho(0,T)$. The values of $(\Delta\rho/\rho)=35\%$, 43%, 38%, and 22% have been observed in samples with $T_A=200\,^\circ$C, 330 °C, 480 °C, and 605 °C, respectively. The data can also be (in fact more commonly are) displayed as $(\Delta\rho/\rho)_{Hs}=[\rho(H,T)-\rho(H_s,T)]/\rho(H_s,T)$, where H_s is the saturation field. The values of $(\Delta\rho/\rho)_{Hs}=54\%$, 75%, 61%, and 28%,

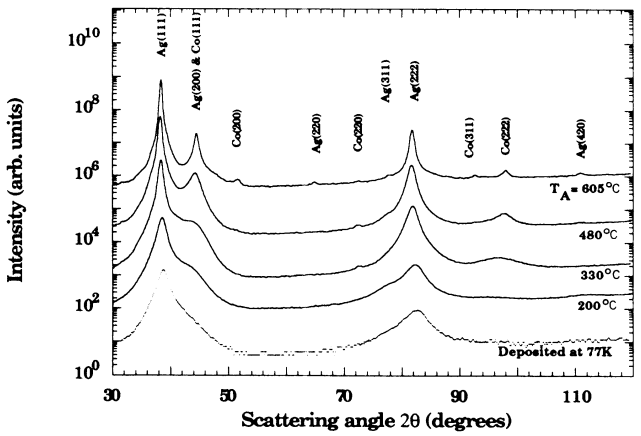

FIG. 1. X-ray diffraction patterns of $Co_{20}Ag_{80}$ sample deposited at 77 K (fcc alloy) and granular fcc Co in fcc Ag after annealing at 200 °C, 330 °C, 480 °C, and 605 °C for 10 min. For clarity, the diffraction patterns have been shifted vertically.

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also shown in Fig. 2 have been observed in samples with $T_A = 200\,^\circ\text{C}$, $330\,^\circ\text{C}$, $480\,^\circ\text{C}$, and $605\,^\circ\text{C}$, respectively. These values are among the largest reported. The field value of H_s changes progressively with T_A; H_s is quite large for the sample annealed at $T_A = 200\,^\circ\text{C}$, becomes smaller for that of $T_A = 330\,^\circ\text{C}$, and has a value of about 10 kOe for those of $T_A = 480\,^\circ\text{C}$ and $605\,^\circ\text{C}$.

The apparent dependence of GMR on the magnetic field is not particularly revealing because it is a direct consequence of the field dependence of the magnetization (M).[9] The magnetization behavior (M vs H) of each sample was measured by a SQUID magnetometer at 5 K. In the original unmagnetized state of a granular magnetic solid, the magnetic axes of the Co particles are randomly orientated with zero net magnetization ($M = 0$), at which $\Delta\rho/\rho = 0$. The largest (most negative) value of $\Delta\rho/\rho$ is achieved when the magnetic axes of all the particles are aligned, i.e., $M = M_s$, where M_s is the saturation magnetization. It is most instructive to plot the GMR as a function of M/M_s, as shown in Fig. 3. Since GMR is an even function of M/M_s, only the results for positive M/M_s are shown. The data of all the samples, regardless of their H_s values, have similar dependence on M/M_s. The fractional GMR data can be described as

$$\frac{\Delta\rho}{\rho} = \frac{\rho(H,T) - \rho(0,T)}{\rho(0,T)} = -AF[(M/M_s)^2] , \qquad (1)$$

where F is a function of the argument $(M/M_s)^2$, and A determines the ultimate size of the GMR. The function $F(x)$ is monotonic, bounded between 0 and 1, with the limiting values of $F(x) \to 0$ at $M \to 0$; and $F(x) \to 1$ at $M \to M_s$. The simplest form of F is $F = (M/M_s)^2$ which describes the data well for small values of $(M/M_s)^2$. It is important to emphasize that in a granular system, all the Co moments within a single-domain Co particle remain ferromagnetically aligned, and M is the *global* magnetization, averaged over many Co particles. The external field only *rotates* the magnetization axes of the Co particles. In other words, in a granular system, $M = \langle \mathbf{M}\cdot\mathbf{H} \rangle / H = M_s \langle \cos\theta \rangle$, where θ is the angle between the magnetization axis of a Co particle and the external field, and $\langle \cos\theta \rangle$ is the average over many Co particles.

Another key feature is that the GMR in granular systems is *isotropic*. In Fe-Cr multilayers,[1] GMR has been measured in all three geometries ($\rho_\perp$, $\rho_\parallel$, and ρ_T). The results for $\rho_\parallel$ and ρ_T are very similar, but very different from that of $\rho_\perp$. In particular, the value of H_s for $\rho_\perp$ is about 20 kOe larger than those of $\rho_\parallel$ and ρ_T, due to the demagnetizing factor ($4\pi M = 20$ kOe for Fe in the layer structure). In a granular system, no such pronounced anisotropy in GMR should be expected. Indeed, as shown in Fig. 2(b), the results for $\rho_\parallel$, ρ_T, and $\rho_\perp$ for the sample with $T_A = 330\,^\circ\text{C}$ are practically the same. The values for $\rho_\parallel$ and ρ_T are identical, and those of $\rho_\perp$ are only slightly larger for $H < H_s$. The small deviation is due to the fact that the Co particles are not perfectly spherical. Therefore, when ferromagnetic alignment of the Co particles is achieved, the GMR is independent of the angle between the magnetization direction and the current.

From the above results, the resistivity of a granular metal exhibiting GMR can be expressed as

$$\rho(H,T) = \rho_0 + \rho_{\text{phonon}}(T) + \rho_m(T)\{1 - F[(M/M_s)^2]\} , \qquad (2)$$

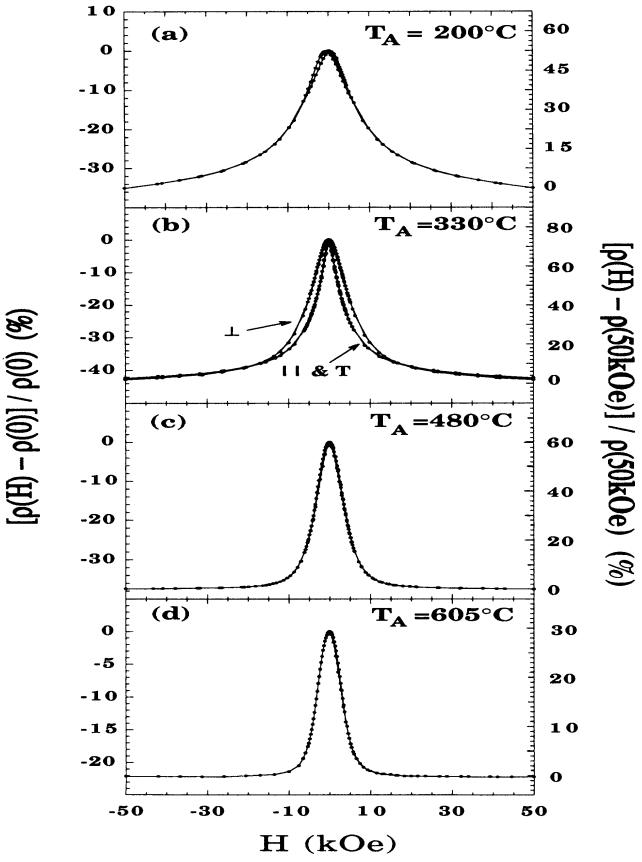

FIG. 2. Perpendicular magnetoresistance for $Co_{20}Ag_{80}$ samples annealed at (a) 200 °C, (b) 330 °C, (c) 480 °C, and (d) 605 °C. For the results in (b), the longitudinal ($\rho_\parallel$) and transverse (ρ_T) are the inner curves which are indistinguishable, whereas the perpendicular ($\rho_\perp$) is the outer curve.

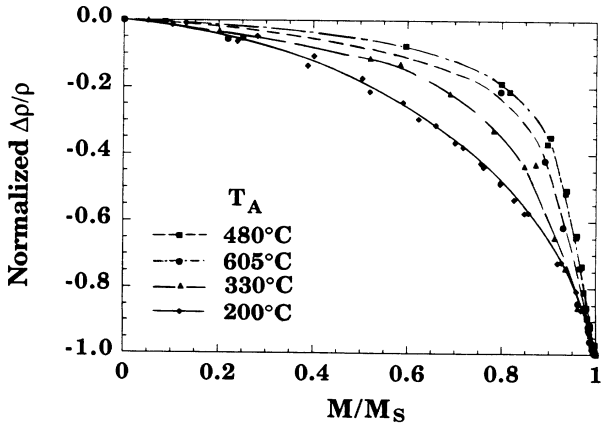

FIG. 3. Normalized perpendicular magnetoresistance $(1/A\,\Delta\rho/\rho)$ vs (M/M_s) of $Co_{20}Ag_{80}$ annealed at 200 °C, 330 °C, 480 °C, and 605 °C. The curve connects the data of each sample.

where ρ_0 is the usual temperature-independent contribution from defects, $\rho_{\text{phonon}}(T)$ is the phonon contribution, quasilinear in T, and $\rho_m(T)$ is the contribution responsible for the GMR. The fractional GMR is

$$-\frac{\rho_m(T)}{\rho_0+\rho_{\text{phonon}}(T)+\rho_m(T)}F[(M/M_s)^2], \quad (3)$$

where the prefactor is just the factor A in Eq. (1). To realize a large value of GMR, $\rho_m(T)$ must be a significant fraction of the total resistivity. It is also important to examine $\rho_m(T)$ itself, which can be deduced from $\rho_m(T)\approx\rho(0,T)-\rho(50\text{ kOe},T)$ for those samples that H_s is less than 50 kOe. The temperature dependence of the total resistivity $\rho(H,T)$ at $H=0$ and 50 kOe, as well as those of GMR and $\rho_m(T)$ are shown in Fig. 4. It is clear that most of the temperature dependence of resistivity is carried by $\rho_{\text{phonon}}(T)$. However, $\rho_m(T)$ is not constant but weakly dependent on T. In fact, $\rho_m(T)$ decreases slightly and roughly linear with T. Both the total resistivity and the value of $\rho_m(T)$ become progressively smaller for samples with higher T_A. For samples with $T_A=330\,^\circ$C, 480 °C, and 605 °C, the values of ρ_m at 5 K are 5.5, 3.5, and 1.6 $\mu\Omega$ cm. It may be noted that in conventional magnetoresistive materials such as Permalloy the anisotropic magnetoresistance is about 0.5 $\mu\Omega$ cm.[14] For increasing T, the magnitude of the fractional GMR has been universally observed to be reducing. For samples with $T_A=330\,^\circ$C and 480 °C, the reduction is caused by the combination of increasing $\rho_{\text{phonon}}(T)$ and decreasing $\rho_m(T)$. For the sample with $T_A=605\,^\circ$C, since $\rho_m(T)$ is practically constant, the decrease of GMR is essentially caused by $\rho_{\text{phonon}}(T)$ alone. For the present samples, the values of $(\Delta\rho/\rho)$ at room temperature are 12%, 17%, 19%, and 5% [$(\Delta\rho/\rho)_{Hs}=14\%$, 20%, 24%, and 5%, respectively] for the samples annealed at 200 °C, 330 °C, 480 °C, and 605 °C, respectively.

The quantity $\rho_m(T)$ due to the scattering of conduction electrons from the ferromagnetic entities, has to be addressed theoretically. A few theoretical models of GMR so far have utilized spin-dependent scattering applied to the multilayer structure.[1,5–7] The present results show that GMR can also be realized in magnetically inhomogeneous media containing nonaligned ferromagnetic entities on a microscopic length scale without requiring a multilayer structure. The quantity $\rho_m(T)$ depends on the size and number of the ferromagnetic entities within the mean-free path (Λ), and the magnetic orientations of the entities are contained in the factor of $[1-F[(M/M_s)^2]$.

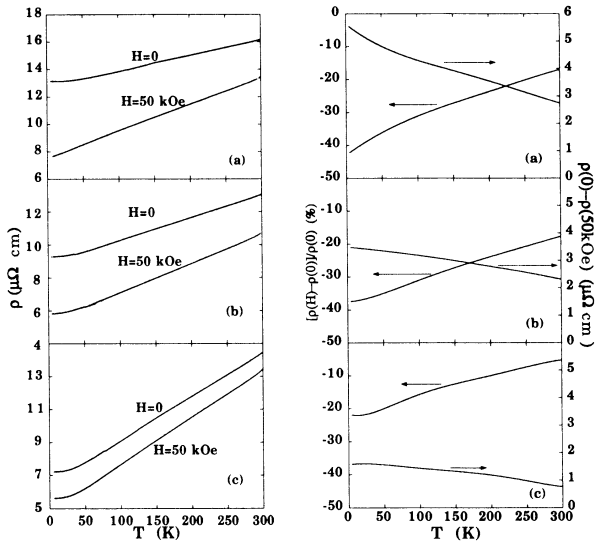

FIG. 4. Temperature dependence of the resistivity at $H=0$ and 50 kOe (left panel), giant magnetoresistance and $\rho(0,T)-\rho(50\text{ kOe},T)\approx\rho_m(T)$ (right panel) of $Co_{20}Ag_{80}$ annealed at (a) 330 °C, (b) 480 °C, and (c) 605 °C.

It indicates that the conduction electrons sense both the magnetic entities *and* the directions of their magnetic axes. The GMR is isotropic, its magnitude is independent of the orientation of M with respect to the current. Within Λ, of the order of a few hundred Å estimated from resistivity, the scattering events due to *disordered spins* contribute to the extra resistivity. The size and the number of the ferromagnetic entities with Λ dictates the ultimate magnitude of GMR. These essential features are to be incorporated into viable models of GMR. As mentioned above, the size of the Co particles increases monotonically with the annealing temperature T_A, but the GMR shows a maximum at about $T_A=330\,^\circ$C as shown in Fig. 2. For larger Co particles resulting from a higher T_A, the scattering events within Λ are fewer, hence a smaller GMR.

Finally, it should be mentioned that we have also observed comparable values of GMR in granular systems of Fe-Cu, Fe-Ag, and several other systems, as well as other compositions of Co-Ag, the results of which will be described elsewhere.

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