Giant magnetoresistance and its evolution in the granular Fe_xAg_{100-x} system ($0 \leqslant x \leqslant 100$)

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We have studied the magnetotransport and magnetic properties of a metallic granular Fe_xAg_{100-x} system, fabricated by using a magnetron cosputtering technique. Giant magnetoresistance was observed in a narrow range between $x=10$ and 25 vol %. Both the Fe volume fraction and the sample orientation significantly influence the magnetoresistance. This granular system also demonstrates some unique features as compared to multilayer systems.

Over the last few years, we have seen intense research activities in the area of magnetotransport in magnetic layered structures, initiated by the discovery of the fascinating giant magnetoresistance (GMR) in Fe-Cr multilayers with antiferromagnetic interlayer coupling.[1] Extensive research efforts have resulted in the finding of many other layered systems,[2–4] e.g., Co-Cr, Fe-Cu, with the GMR effect. Due to its fundamental significance, this effect has also received concurrent theoretical attention. Various models have been proposed to explain the GMR.[5–7] However, the mechanism remains far from well understood. In the application front, the study of GMR is equally significant since it promises a new generation of magnetic sensors and recording devices. Recently, a stunning development has introduced a new dimension to the research of GMR. It has been reported[8,9] that GMR also occurs in ferromagnetic granular materials in which magnetic single-domain Co particles are embedded in a metallic Cu matrix. Multilayer structures are no longer a prerequisite for the exhibition of GMR. This report not only offers new challenges for the fundamental understanding of the mechanism, it is also noteworthy in application because granular materials can be easily fabricated over large areas.

The demonstration of GMR in a nonlayered system[8,9] raises many new questions pertinent to the basic study and application of that effect, among them the diversity of GMR in different granular materials, the effective fabrication method to yield granular systems with configuration optimal to GMR, the role of material parameters such as particle volume fraction and size on GMR. To investigate some of these questions, we have fabricated and studied a new granular system, Fe_xAg_{100-x}, over the whole volume fraction (x) range. In this letter, we report the observation of GMR and its uniqueness in Fe-Ag granular solid. The GMR is found to be very sensitive to the Fe volume fraction and it disappears in both the Fe-poor and the Fe-rich regions of the system. We will also present magnetization measurement related to the GMR effect.

Traditionally, magnetic granular solids are made in the forms of cermets[10] (particles embedded in a ceramic matrix) or free-standing particles. The fabrication of metallic granular materials has gained some attention only recently.[11] Childress and Chien[11] have taken advantage of the fact that certain binary metallic elements are immiscible in the equilibrium state. Using an effective vapor quenching technique, they have made metastable homogeneous alloys and then annealed the samples at an elevated temperature to induce phase separation. As a result, ultrafine metallic particles, 1–20 nm in size, coexist with a phase-pure metallic matrix. The GMR effect in the granular system was discovered in samples fabricated in this fashion.[8,9] It should be pointed out that the number of metallic elements, that are mutually insoluble with magnetic transition elements, Fe, Co, and Ni, is rather limited. Among the few, we have chosen the Fe-Ag system. It is known that the mutual solubility of Fe and Ag is negligible (<1 ppm)[12] at ambient temperature. There are no line compounds or alloys in the equilibrium Fe-Ag phase diagram.[12] To the best of our knowledge, GMR has not been reported in Fe-Ag multilayers.

Our samples were fabricated by using a magnetron cosputtering technique. The high vacuum system is equipped with a cluster of three magnetron guns aimed at the same spot. Two of these guns were loaded with pure Fe and Ag targets ($\geqslant$99.9%) that were cosputtered. Before each run, the background vacuum was about 1×10^{-7} Torr. The Ar sputtering gas pressure was kept at 4 mTorr by a gas flow controller. All samples were deposited onto Si wafers at ambient temperature, with no post-deposition annealing. We used the standard photolithography and wet etching technique to pattern samples for resistivity measurement. The structure and phase formation was checked by using x-ray and electron diffraction, as well as transmission electron microscope (TEM). A superconducting quantum interface device magnetometer was employed to measure the magnetization (M) of these samples.

In multilayer systems, GMR is very sensitive to material parameters. This is also expected in granular systems. We have measured magnetoresistance (MR) at $T=4.2$ K, where phonon or magnon contributions to resistivity ρ are negligible. Shown in Fig. 1 is the MR, $\Delta\rho/\rho$ (referred to the zero field ρ), obtained with two H orientations ($\parallel$ and $\perp$ film) for five representative samples. The value of ρ attains a maximum whenever the magnetization $M=0$. As H is increased, ρ decreases together with the gradual alignment of the magnetic moments. The magnitude of $\Delta\rho/\rho$ in the field range of $0-\pm8T$ changes considerably from sample to sample. Figure 2 depicts $\Delta\rho(8T)/\rho$ and ρ_0 at zero

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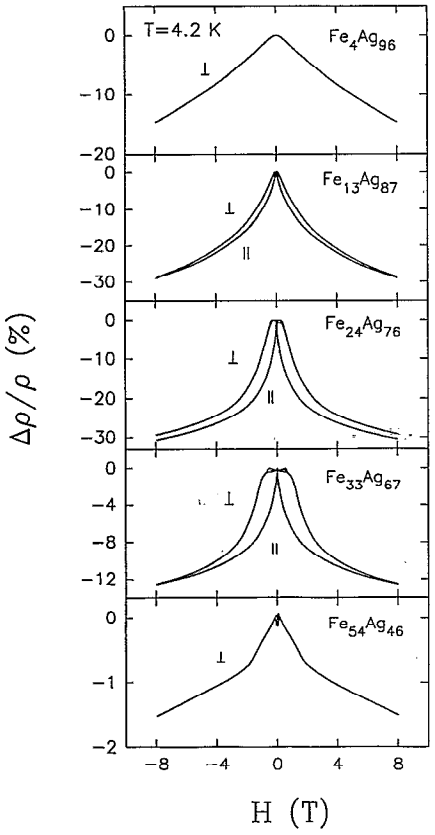

FIG. 1. Magnetoresistance vs in-plane (∥) and out-plane (⊥) magnetic field for five representative Fe_xAg_{100-x} samples at $T=4.2$ K.

field as functions of Fe volume fraction x. Very interestingly, $\Delta\rho/\rho$ increases abruptly with x in the Fe-poor region, then $\Delta\rho/\rho$ reaches a maximum plateau with $\Delta\rho/\rho \approx 30\%$ in the region of $10\% \leqslant x \leqslant 25\%$. Beyond $x=25\%$, $\Delta\rho/\rho$ decreases gradually with x into the Fe-rich region. The large $\Delta\rho/\rho$ observed in the plateau region is about the same as that in the Co-Cu[8,9] and the Co-Ag[13,14] granular systems, and is as large as some multilayer systems with

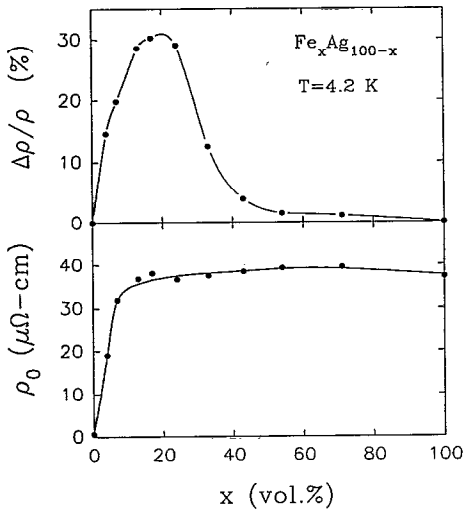

FIG. 2. Dependence of magnetoresistance $\Delta\rho/\rho$ and ρ_0 at zero field on Fe volume fraction at $T=4.2$ K.

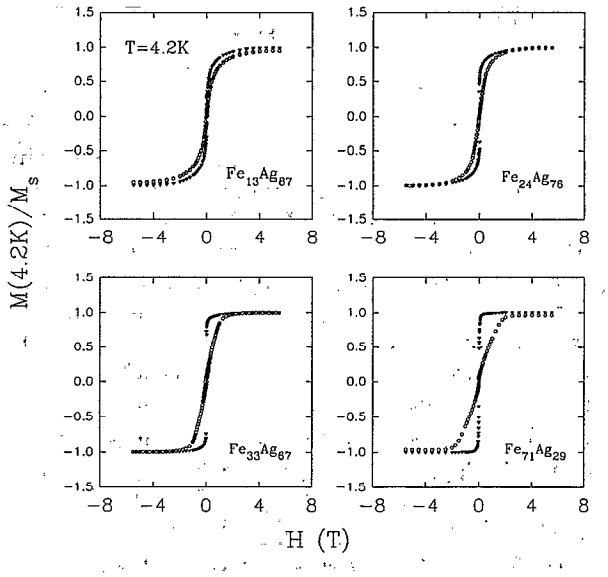

FIG. 3. Perpendicular and parallel magnetic hysteresis curves for four representative Fe_xAg_{100-x} samples at $T=4.2$ K. The curves with lower saturation field are obtained in a parallel magnetic field.

the largest GMR.[1-4] We note that the reported $\Delta\rho/\rho$ is generally referenced to the saturation value of ρ at high fields, whereas we use the maximum ρ_0 at zero field as a reference. $\Delta\rho/\rho$ would exceed 45% in our system if we use the common definition of $\Delta\rho/\rho$.

X-ray and electron diffraction patterns revealed that the Fe-Ag system in the GMR peak region ($x<30\%$) has a fcc structure with lattice constant almost identical to that of pure Ag (with less than 2% variation). This is an indication that uniform Fe-Ag alloy is not formed because, otherwise, one would expect to observe a variation of lattice constant with Fe content. Direct revelation of small Fe clusters is nearly impossible in diffraction study for two reasons. First, the spectral lines of the bcc Fe-phase overlap with those of fcc Ag. Second, the atomic number Z of Fe is much smaller than that of Ag, causing the signal from the Fe phase difficult to be detected in Fe-poor region. However, TEM results on samples with optimal GMR always showed the existence of very fine grains (~20 Å) uniformly dispersed in a sample.

Figure 3 shows the magnetization curves at $T=4.2$ K for four representative samples with H perpendicular and parallel to the sample plane. We found that all samples including Fe_4Ag_{96} are ferromagnetic at $T=4.2$ K and the magnetization of the Fe component is equal to that of the bulk Fe within our experimental error. This indicates that the phase separation between Fe and Ag is complete. The anisotropy in M as seen in Fig. 3 is due to the shape anisotropy of the Fe particles in the Fe-poor region and due to the thin-film-shape anisotropy in the Fe-rich region where particle percolation occurs. In all of the samples, magnetic hysteresis is rather small with H_c about a few tens of Gauss.

An inspection of Figs. 1 and 3 shows that the anisotropy between the longitudinal and perpendicular MR is closely related to the anisotropy in M. When H is perpendicular to a sample plane, a large H is required to saturate

M, and MR decreases with H slowly. On the other hand, when H is in the parallel orientation, M tends to be saturated easily, and MR also decreases with H with a much larger initial slope. In application, the initial slope of MR is most important. A large slope will enhance the sensitivity of a magnetic sensor particularly at low H.

It is commonly held that GMR in layered structures is caused by spin-dependent scatterings[5–7] between the conduction electrons and the magnetic entities (interface and/or bulk). The effective electron scattering rate $1/\tau(H)$ depends on the relative orientations of M in each layer. Both experiments[1,2] and theoretical models[6,7] have shown that $1/\tau(H)$ approaches a minimum as M saturates above a critical H. In some granular systems, it is shown that $\Delta\rho/\rho \propto -[M(H)/M_s]^2$, where M_s is the saturated magnetization.[9] In all of these cases, $\Delta\rho/\rho$ becomes H independent when M reaches M_s. This behavior is certainly not observed in our Fe-Ag granular system. As can be seen in Figs. 1 and 3, the saturation of M does not guarantee $\Delta\rho/\rho$ to reach its minimum. In fact, M is saturated with only a few hundred Gausses in some samples in the parallel configuration, but $\Delta\rho/\rho$ shows no sign of saturation even at $H=\pm 8T$, our maximum field. This is a unique feature of GMR in this granular system, which needs to be reckoned with in theoretical models.

The evolution of GMR with Fe volume fraction as shown in Fig. 2 is another important feature of this system. The maximum GMR is achieved in a moderate volume fraction range (10%–25%). The suppression of GMR in the Fe-poor region results from a subdued magnetic scattering due to the low concentrations of Fe particles. This effect is also evident in the residual resistivity ρ_0 shown in Fig. 2. As the GMR effect reduces with decreasing x, so does ρ_0 which includes both magnetic scatterings and disorder scatterings. Above $x=25\%$, the weakening of GMR is likely due to the clustering of Fe particles. As x increases, the percolation effects sets in. The clustering of particles has the effect of enlarging the effective particle size. A larger particle has a smaller surface area to volume ratio, which affects GMR adversely because interfacial spin-dependent scattering is crucial to GMR.[5–7] A quantitative analysis of the volume-fraction dependence of GMR is difficult at this stage because of the lack of an understanding of the GMR mechanism in granular structures. Nevertheless, the evolution of GMR shown in Fig. 3 reveals the strong sensitivity of GMR to the material configuration, this fact should be an essential ingredient in theories.

In summary, GMR and its variation with volume fraction have been observed in a new Fe-Ag granular system. The maximum GMR occurs in a range of $10\leqslant x\leqslant 25\%$, while in both Fe-poor and Fe-rich regions GMR is substantially suppressed. Contrary to multilayer systems, MR in the granular Fe-Ag system remains unsaturated up to a high field of ± 8 T, even though magnetization is saturated at a much lower field. The present study shows that GMR is a rather commonly occurred phenomenon in magnetic granular systems, which already rival the best magnetic multilayers in terms of the magnitude of GMR and the ease in material fabrication.

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