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Tunneling giant magnetoresistance in coevaporated $\text{Fe}_x(\text{SiO})_{1-x}$ thin films

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$\text{Fe}_x(\text{SiO})_{1-x}$ thin films, with volume fraction x ranging from 0.04 to 0.6, have been prepared by coevaporation. Magnetization, resistivity, and magnetoresistance measurements show that the behavior of this composite is metallic like for $x \geq 0.35$. For $0.2 \leq x \leq 0.27$, the confrontation of all measurements prove the existence of a tunneling giant magnetoresistance between ferromagnetic grains at $T < 300$ K, where the mean radius of the grains is estimated to be about 1 nm. © 2000 American Institute of Physics. [S0021-8979(00)05923-5]

Correlation between magnetoresistance and superparamagnetism was first shown in early work by Gittleman *et al.*¹ in granular Ni–SiO₂. The recent discovery of the so-called giant magnetoresistance in inhomogeneous composites made of nanoscale ferromagnetic granules (Fe, Co, Ni) embedded in a metallic host (Ag, Cu) has stimulated new studies with the insulating host^{2–6} (Al₂O₃, SiO₂). They are generally prepared by magnetron sputtering. Our group^{7,8} has recently studied the structure and optical properties of silicon clusters in amorphous SiO_x and SiO_x:H thin films prepared by evaporation, and the purpose of this communication is to show that the evaporation technique also permits us to obtain nanometric magnetic grains in an insulating host which present the giant magnetoresistance effect.

The $\text{Fe}_x(\text{SiO})_{1-x}$ alloys were obtained by coevaporation of Fe and SiO from an electron gun and a thermal cell, respectively. By varying the deposition rates of Fe and SiO, we prepared $\text{Fe}_x(\text{SiO})_{1-x}$ samples with different *volume fractions* x ranging from 0.04 to 0.6. The substrates were maintained at 100 °C. The total theoretical thickness, calculated with the density of Fe and SiO, was 50 nm for the transmission electron microscopy (TEM) and 200 nm for the other experiments. Classical TEM and diffraction experiments show that the samples seem homogeneous and amorphous for x less than 0.35. The metallic clusters are absent or too small to be seen by this technique. The $\text{Fe}_x(\text{SiO})_{1-x}$ samples are crystallized for $x \geq 0.45$. The diffraction spectrum shows crystals with the bcc structure of pure iron. For electrical and magnetic measurements, the samples were deposited onto float glass and silicon substrates, respectively. The conductivity has been measured from 77 to 300 K. Magnetoresistance has been measured with a current in the plane of the film in a superconducting coil equipped cryostat with a maximum field of 70 kOe. The magnetic properties were determined with a semiconducting quantum interference device (SQUID) magnetometer.

The low field ($H = 20$ Oe) magnetization has been measured after field cooling (FC) or zero FC (ZFC) as a function of temperature for all samples. A magnetic signal can be

observed in these conditions only for $x \geq 0.2$. The FC and ZFC curves are characteristic of granular systems for $0.2 \leq x \leq 0.27$ as shown in Fig. 1(a) for $x = 0.2$. They exhibit a $1/T$ slope at high temperature and a narrow peak of the ZFC curve for the so called blocking temperature T_B : T_B is equal to 9 K for $x = 0.2$ and 15 K for $x = 0.27$. For $x = 0.35$ [Fig. 1(b)], no $1/T$ slope is observed. The decrease of magnetization with increasing temperature rather remains the behavior of a homogeneous ferromagnetic material. The low temperature magnetization is close to bcc iron, although no crystallized phase has been observed in the TEM experiments. We note that a hysteresis between the FC and ZFC appears below $T = 25$ K, and this may be attributed to some isolated magnetic grains. The FC and ZFC curves for $x = 0.45$ correspond to a homogeneous soft ferromagnetic material without hysteresis between the FC and ZFC curves at low temperatures.

High field measurements have allowed us to detect a magnetic signal at 5 K for $x = 0.13$. This indicates the existence of some magnetic grains with a strongly reduced magnetization ($0.25\mu_B/\text{Fe}$ atom). The fit of the magnetization curve with a Langevin function provides a particle moment of $53\mu_B$, which corresponds to the radius of a particle of 8 Å within the assumption of a magnetization of $0.25\mu_B/\text{Fe}$ atom. The field dependence of magnetization is plotted in Fig. 2 for $x = 0.2$. We first note that the magnetization is reduced by a factor 2 with respect to bulk bcc iron, suggesting that some iron may be dispersed in the matrix or that the magnetic granules may be amorphous. The magnetization curves show a Langevin like behavior for $T \geq 100$ K, well above the blocking temperature, in agreement with the FC and ZFC curves. Nevertheless, the fit with a unique Langevin function presents some discrepancies with the experimental points, which is readily attributed to the existence of a distribution of size. Within the assumption of a simple normal distribution of radius R : $f(R) = 1/R$, the sum of the Langevin functions is then analytic and can be written as

$$M(T, H) = \frac{M_s(T)}{(\mu_2 - \mu_1)x} \ln \left(\frac{\mu_1 \text{sh}(\mu_2 x)}{\mu_2 \text{sh}(\mu_1 x)} \right), \quad (1)$$

where $x = H/kT$, $M_s(T)$ is the saturation magnetization, and μ_1 (respectively, μ_2) is the minimum (respectively maxi-

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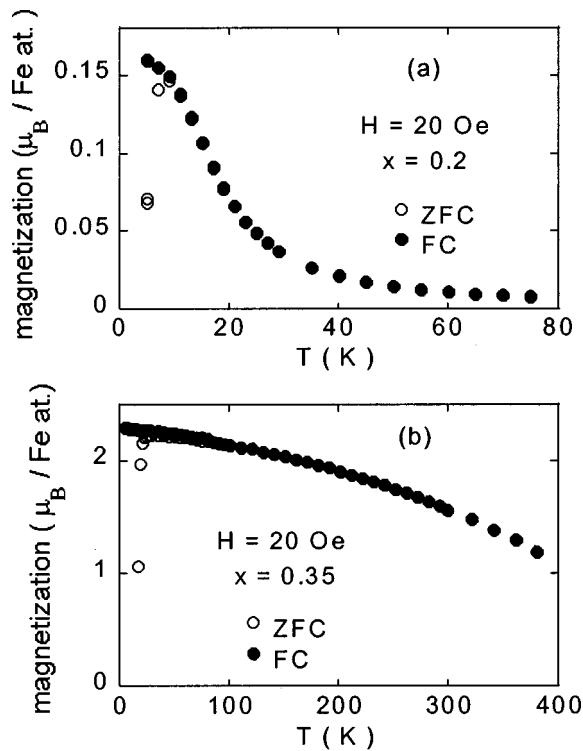


FIG. 1. Low field ($H=20$ Oe) magnetization measured as a function of temperature after field cooling (FC) or zero field cooling (ZFC) for $\text{Fe}_{0.2}\text{Si}_{0.8}$ (a) and $\text{Fe}_{0.35}\text{Si}_{0.65}$ (b).

num) value of the moments of the particles in the distribution. The fit of the magnetization curve at $T=100$ K with expression (1) provides $\mu_1=60\mu_B$ and $\mu_2=1300\mu_B$. Assuming that the magnetization of the grains is given by $M_s(T)$ and that their density corresponds to the density of bulk iron, we can evaluate the minimum and maximum radii of the distribution: $R_1=5$ Å and $R_2=14$ Å. The mean radius of the distribution is then equal to 9 Å. The same treatment for $x=0.27$ provides a distribution of moments of particles which spreads from $70\mu_B$ to $2600\mu_B$, which corresponds to a radius of 6 and 19 Å with a mean size of 11 Å. We note that the moments of the particles are much lower than those obtained with $\text{Fe}(\text{SiO}_2)$ for the same volume fraction.² For

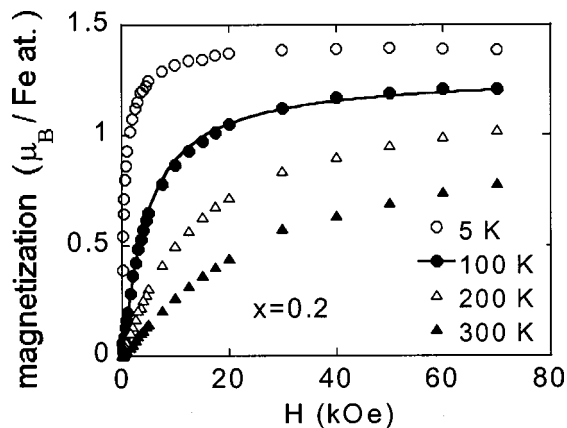


FIG. 2. Magnetization as a function of applied field for $\text{Fe}_{0.2}\text{Si}_{0.8}$ composites. The solid line represents a fit with a sum of Langevin functions.

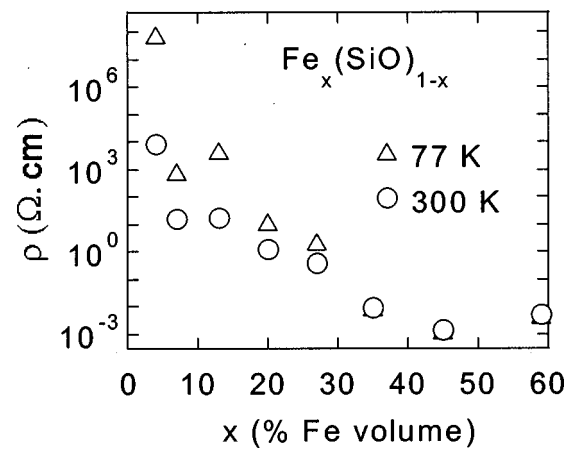


FIG. 3. Resistivity ρ of $\text{Fe}_x(\text{SiO})_{1-x}$ measured at room temperature and at 77 K as a function of x .

$x \geq 0.35$, the measurements are analogous to those of a ferromagnetic material. In conclusion, magnetization measurements show superparamagnetism in coevaporated $\text{Fe}_x\text{SiO}_{1-x}$ for $0.13 \leq x \leq 0.27$.

Figure 3 shows the electrical resistivity ρ of $\text{Fe}_x(\text{SiO})_{1-x}$ films at room temperature and at 77 K. The values are similar to those obtained in the granular Ni-SiO₂^{1,9} and Fe-SiO₂^{2,10} systems. There is an abrupt increase in ρ with SiO content around $x=0.3$, which corresponds to a change of mechanism of electrical conductivity. For $x > 0.3$, the temperature coefficient of resistivity (TCR) is positive, indicating a metallic conductivity, while for $x < 0.3$ it becomes negative, suggesting a nonmetallic conductivity such as semiconductorlike or tunneling conductivity. An interesting observation is that the value of conductivity at the composition where TCR vanishes is in the range of $10^2-10^3 \Omega^{-1} \text{cm}^{-1}$. This value of minimum metallic conductivity agrees with Mott's prediction of the limiting conductivity of a metal. We remark that this metallic percolation coincides with the crossover between the superparamagnetic and ferromagnetic behaviors. Moreover, it occurs at a volume fraction close to the theoretical value predicted by Landauer¹¹ and is lower than in other composites obtained by sputtering.^{2,4,6,9} Electrical conduction in the dielectric regime of granular metals results from transport of electrons and holes by tunneling from one isolated metallic grain to the next. The temperature dependence of ρ is given by $\rho = \rho_0 \exp(2\sqrt{c}/kT)$, where ρ_0 is a constant and c is the activation energy proportional to both the tunnel barrier thickness and the charging energy of the metallic grain. Figure 4 shows that this behavior is observed between 77 and 300 K for the samples with $x < 0.3$. It was also verified that these curves are readily distinguishable from the Mott $\ln \rho \sim 1/\sqrt{T}$ or the $\ln \rho \sim 1/\sqrt{T}$ behavior. The coefficients c decreases from 0.58 eV for $\text{Fe}_{0.04}(\text{SiO})_{0.96}$ to 0.05 eV for $\text{Fe}_{0.27}(\text{SiO})_{0.73}$. These values agree well with the values for Ni-SiO₂,⁹ Pt-SiO₂,⁹ Co-SiO₂,¹¹ and Fe-SiO₂ systems.²

The magnetoresistance, $\text{MR}(H) = [\rho(H) - \rho(0)]/\rho(0)$ is shown in Figs. 5(a) and 5(b) as a function of the applied field for $x=0.2$ and $x=0.35$ at $T=100$ K. This measurement has been made with a constant current parallel to the applied

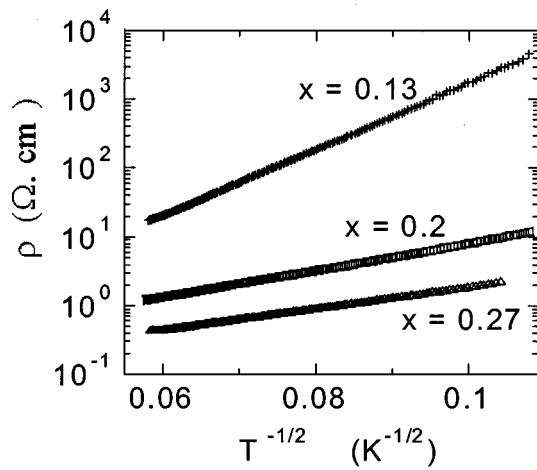


FIG. 4. Electrical resistivity in a logarithmic scale as a function of $T^{-1/2}$ for different $\text{Fe}_x(\text{SiO})_{1-x}$ samples.

field. A decrease of the resistance with applied field, which is the signature of a giant magnetoresistance (GMR) effect in this geometry, is clearly shown for $x=0.2$. The same feature has been observed for $x=0.27$. No GMR effect has been clearly detected at 300 K. For these concentrations, the temperature dependence of the resistivity is consistent with a tunneling effect between isolated grains; this GMR effect is then attributed to a spin dependent tunneling conduction between ferromagnetic grains. Assuming that this effect is pro-

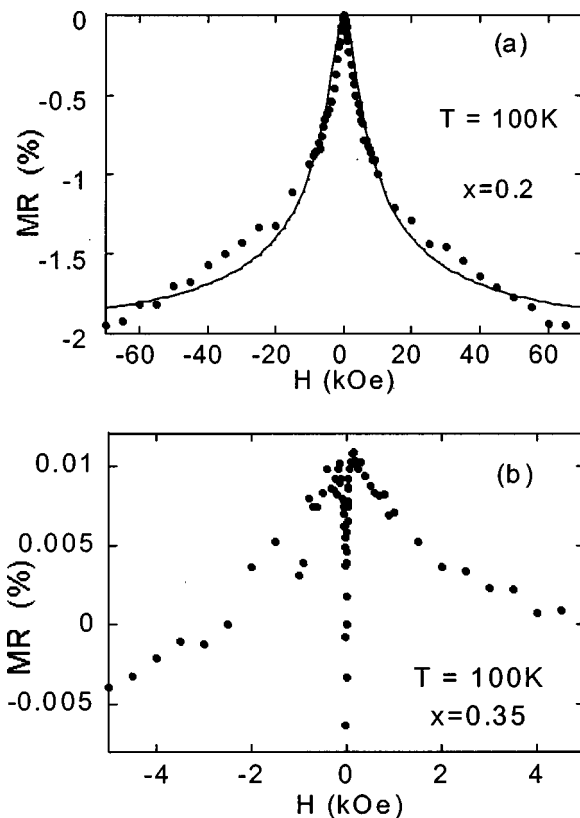


FIG. 5. Magnetoresistance as a function of applied field at $T=100$ K for $\text{Fe}_{0.2}\text{SiO}_{0.8}$ (a) and $\text{Fe}_{0.35}\text{SiO}_{0.65}$ (b). The solid line for $x=0.2$ represents a fit with a term proportional to the squared magnetization.

portional to the square of magnetization, the $\text{MR}(H)$ curve for $x=0.2$ has been fitted by a term proportional to $M^2(H, T)$ [Eq. (1)] with the parameters obtained from the magnetization experiments. A quite good agreement with the experimental points is obtained, which confirms that the MR effect is due to the particle size distribution determined above. The rather low particle moments of this distribution explains the absence of detectable MR signal at ambient temperature and of saturation at high fields. As a consequence, the maximum tunneling GMR effect (2% at 100 K for $x=0.2$) remains weak compared to the results of other groups.^{2-4,6} For $x=0.35$, the MR presents a minimum at low fields ($H < 300$ Oe) and a decrease of MR for high fields ($H > 300$ Oe). The existence of both these effects may be attributed to a large distribution of size of particles: the larger grains giving rise to an anisotropic magnetoresistance (AMR) at low fields, whereas the smaller produce GMR for higher fields. This agrees with the features of the FC-ZFC curve. As the temperature coefficient of resistivity is positive for this sample, the GMR effect can be rather attributed to a usual metallic conduction between magnetic grains. For $x=0.45$, only an AMR effect is observed, which is characteristic of a homogeneous ferromagnetic material.

Magnetization, resistivity, and magnetoresistance measurements have been performed with $\text{Fe}_x(\text{SiO})_{1-x}$ coevaporated composites with volume fraction x ranging from 0.04 to 0.6. For $x \geq 0.35$, it behaves like a metallic system with a magnetization per iron volume closed to pure iron. It exhibits both anisotropic and GMR effect for $x=0.35$ as an inhomogeneous metallic system, whereas only an anisotropic magnetoresistance is observed for $x \geq 0.45$. For $x \leq 0.27$, its behavior is semiconducting or insulatinglike, with a strongly reduced magnetization per iron volume compared to bulk iron. For $0.2 \leq x \leq 0.27$, all measurements prove the existence of a tunneling GMR at low temperature ($T < 300$ K) between ferromagnetic grains. Their mean radius has been evaluated to be about 1 nm. Nevertheless, the reduced magnetization and the absence of crystallites in TEM measurements suggest that the magnetic granules are badly crystallized or amorphous. Some experiments with annealed samples are planned to study the correlation of the structure and transport properties of these composites.

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