Magnetoresistance of granular ferromagnets

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We report a comparative study of the magnetic and magnetoresistance phenomena in two types of granular ferromagnets: ferromagnet-normal-metal mixtures Ni-Ag, Co-Ag, and ferromagnet-insulator mixtures Ni-SiO_2, Co-SiO_2. Anisotropic magnetoresistance and giant magnetoresistance (GMR) are identified in both types of systems above and below the magnetic component percolation threshold respectively. The GMR effect in granular ferromagnets is shown to be essentially independent of the nature and resistance of the nonmagnetic intergranular matrix. [S0163-1829(97)04010-1]

INTRODUCTION

Granular ferromagnets are immiscible mixtures of two materials one of which is ferromagnetic in the form of nanoscopic-size polycrystalline grains and the other is nonmagnetic. The nonmagnetic material can be either metallic (Ag, Au, Cu, . . .) or insulating (SiO_2, Al_2O_3, . . .) in crystalline or amorphous form. In the forthcoming discussion we shall call the ferromagnet-normal-metal granular mixtures as FM and ferromagnet-insulator ones FI respectively.

Magnetic properties of granular ferromagnets are thoroughly studied[1] and known to depend mainly on the topological structure of the ferromagnetic component. When the size of individual grains is sufficiently large for the existence of ferromagnetism, the isolated nanometer-size ferromagnetic grains are single domains. At low concentration of the ferromagnetic material individual grains are dispersed in a nonmagnetic matrix. When the space orientation of granular easy magnetic axes is random, the system behaves like a superparamagnet.[2] At high magnetic material volume fraction, the magnetic particles coalesce and form a connected network. In this regime, the magnetic phase percolates throughout the material and the sample appears to be a normal ferromagnet. The transition from superparamagnetic to ferromagnetic state seems to be a general property of granular ferromagnets and was found either in ferromagnet-normal-metal (FM) (Ref. 1) and ferromagnet-insulator (FI) (Refs. 1 and 3) mixtures.

Although the magnetic properties of both types of granular ferromagnets are similar, the electronic transport properties are different, especially at low ferromagnet component concentrations. The electric conduction mechanism in all-metallic granular composites does not change much as a function of its composition. The resistivity of a granular mixture can be a few times higher than the resistivity of the pure components[4] due to electron scattering from the grain boundaries. However, the total resistivity is low and it has a positive temperature coefficient over entire concentration range. In granular metal-insulator composites there are two regimes of electronic conduction, depending on the metal volume fraction x. When x is large, the metallic grains coalesce and form a connected metallic network, so that electrons can percolate along the connected metallic channels. This regime is usually called metallic, although the properties which depend on the electron's mean-free path are strongly modified due to strong electron scattering from the dielectric inclusions and grain boundaries. For example, the electrical conductivity decreases by orders of magnitude from its crystalline value, and the temperature coefficient of resistivity, although positive, is very much smaller than in pure metals.[5] When x is small, the metal grains form isolated dispersions in an insulating matrix. Electrical conduction in this dielectric regime is by a hopping mechanism in which the charge carriers are transported from one grain to another via thermally activated tunneling. Transition from the metallic to the dielectric regime is characterized by a percolation threshold, at which the metallic network first becomes disconnected. The temperature coefficient of resistivity changes sign to a negative at the composition and temperature where the contribution to electrical conductivity due to thermally activated tunneling becomes comparable to the contribution along the percolating metallic channels. Below the percolation threshold x_c, the hopping process alone accounts for the conductivity of granular metals.

The first experimental and theoretical studies of the electronic transport properties of granular metals[6] and, in particular, granular ferromagnets[7,8] were performed and published more than two decades ago. Renewed interest in granular ferromagnets has been triggered by the recent discovery of the so-called ''giant magnetoresistance effect'' (GMR) in magnetic multilayers[9–12] and heterogeneous magnetic alloys with ferromagnetic grains embedded in a nonmagnetic metallic matrix.[13,14] It was found that the resistance can exhibit large changes of the same order of magnitude as the resistance itself upon the application of an external magnetic field. Till recently the effort was concentrated on ferromagnetic-normal-metal combinations only. Ferromagnet-insulator systems were neglected, probably because

of the assumption that the resistance of the FI system below the metallic percolation threshold is too high to be measured. Recently, it has been demonstrated[15,16] that FI mixtures can be measured safely in a certain concentration range below the percolation threshold and behavior similar to GMR was rediscovered[7] in a number of ferromagnet-insulator mixtures. Our preliminary[16] comparison of GMR-like magnetotransport in FM and FI systems revealed a surprising qualitative and quantitative similarity in the field-dependent effects in both types of systems.

In this paper we present the results of an extended comparative study of magnetoresistive and magnetic properties of some FM and FI granular ferromagnets.

EXPERIMENTAL RESULTS AND DISCUSSION

Sample preparation

Thin granular films of Ni-SiO_2, Co-SiO_2, Ni-Ag, and Co-Ag were prepared by coevaporation of the starting materials using two independent electron beam guns. The results presented in this paper were obtained on samples deposited on a room-temperature glass substrate without any post-deposition thermal processing. The deposition rate and the relative volume concentration of the two components were monitored and controlled by two quartz thickness monitors. Usually, a set of up to twenty four samples was deposited simultaneously. The relative concentration of the components varied smoothly due to a shift in the geometrical location of the substrate relative to the evaporation sources. Samples with Ni (Co) volume concentrations from 30 up to 100 % were prepared. The absolute concentration accuracy defined by the deposition rate control is about $\pm 5\%$. The relative concentration accuracy of the samples of the same series, as defined by their geometrical location, is better than $\pm 0.3\%$. The films had a strip form with linear dimensions of about 8×2 mm. Most of the results reported here were obtained on 100-nm-thick films. Magnetization measurements were performed on 450-nm-thick samples. 15–30 nm thick films were used for transmission electron microscopy.

Magnetoresistance

Ferromagnet-normal metal mixtures

Two types of magnetoresistive behavior are known in metallic ferromagnetic materials, usually referred as ''anisotropic magnetoresistance'' (AMR) and ''giant magnetoresistance'' (GMR). Anisotropic magnetoresistance[17] is observed in ferromagnetic metals such as Fe, Co, and Ni in alloys (permalloy). Phenomenologically the behavior can be identified by a few characteristic features: (i) it is a property of a bulk ferromagnet; (ii) for a uniform magnetization M, the longitudal resistivity is given by

$$\rho=\rho_{\perp}\sin^2\theta+\rho_{\parallel}\cos^2\theta, \tag{1}$$

where $\rho_{\perp}$ and $\rho_{\parallel}$ are resistivities for current flow perpendicular and parallel (or antiparallel) to the magnetization direction respectively, and θ is the angle between the magnetization and the current. $\Delta\rho/\rho=(\rho_{\parallel}-\rho_{\perp})/\rho_{\perp}$ is of the order of 2% in Ni at room temperature. The effect increases at low temperatures and can reach 15% at 5 K in permalloy. (iii) $\rho_{\parallel}$ is generally (but not always) greater than $\rho_{\perp}$; (iv) magnetic fields needed to observe the effect are of the order of 10–100 G.

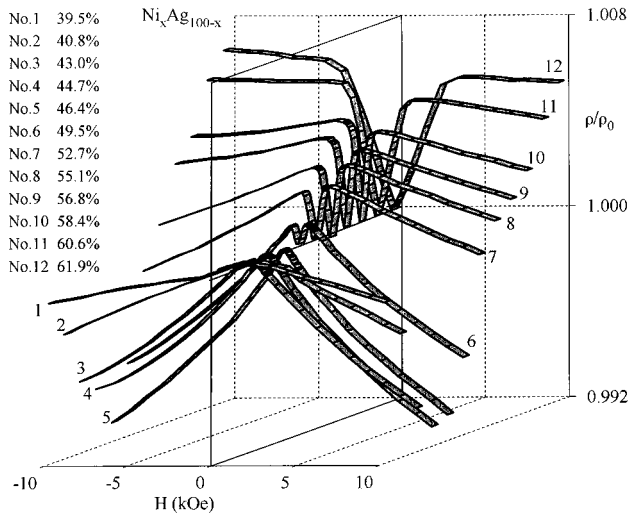

FIG. 1. Normalized room-temperature magnetoresistance of Ni-Ag granular ferromagnets for different Ni volume concentrations x. Magnetic field is applied parallel to the current flow. ρ_0 is the zero field resistivity.

Contrary to the anisotropic magnetoresistance, the GMR effect[18] in heterogeneous ferromagnet-normal-metal systems occurs not within the bulk but in the boundary regions separating magnetic and nonmagnetic materials. The characteristic phenomenological features of GMR are (i) the magnetoresistance is always negative; (ii) in isotropic granular three-dimensional (3D) alloys the effect does not depend on the relative orientation of magnetization with respect to the current flow.

We show in Fig. 1 the magnetic field dependence of the electrical resistivity of Ni-Ag films as a function of Ni volume concentration x with field applied parallel to the current flow. The resistivity data are normalized to the zero field value. Two distinct behaviors can be pointed out. At Ni concentrations up to about 48% only a negative magnetoresistance has been observed. At higher concentrations an additional feature develops: magnetoresistance is sharply positive at low fields, then changes sign and becomes negative. A characteristic ''wing'' curve is developed. With further increase of Ni concentration the positive stroke of resistance progressively increases whereas the negative magnetoresistance slope at higher fields approaches zero.

As has been reported in Ref. 16, the positive ''wing stroke'' in Ni-rich samples ($x>49\%$) disappears when magnetic field is applied perpendicular to the current direction. Contrary to that, the negative magnetoresistance observed in samples with low Ni concentration does not depend on the relative direction between the applied field and the current.[16]

We can, therefore, identify the field-orientation dependent magnetoresistance observed in Ni-Ag at Ni concentrations above 49% as the anisotropic magnetoresistance effect (AMR). Negative isotropic magnetoresistance in samples with low Ni concentration is identified as the giant magnetoresistance (GMR) effect, although the magnitude of the effect in these samples is far from being ''giant.'' Both AMR

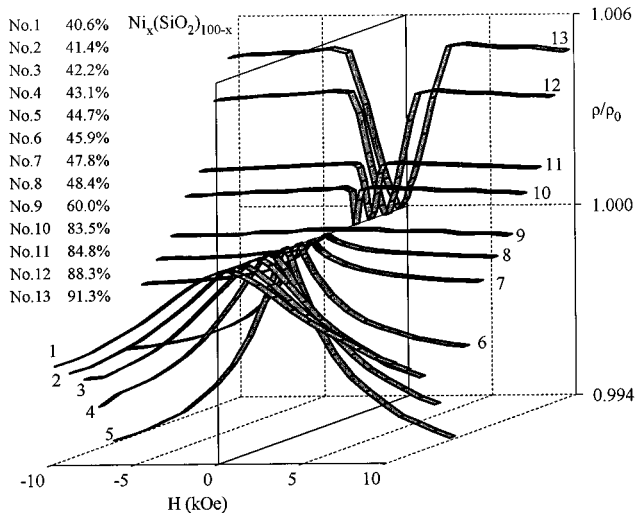

FIG. 2. Normalized room-temperature magnetoresistance of Ni-SiO_2 granular ferromagnets for different Ni volume concentrations x. Magnetic field is applied parallel to the current flow. ρ_0 is the zero field resistivity.

and GMR coexist in samples with intermediate concentrations (see Fig. 1). This behavior is typical and was reproduced in Co-Ag mixtures as well.

Ferromagnet-insulator mixtures

Magnetoresistance of Ni-SiO_2 films for different Ni concentrations with field applied parallel to the current flow is shown in Fig. 2. As in Ni-Ag films (see Fig. 1), a negative magnetoresistance is found in samples with Ni concentrations below 49%. At high Ni concentrations a positive ''wing'' feature is observed when the magnetic field is applied parallel to the current direction. Also,[16] as Ni-Ag, the negative magnetoresistance at low Ni concentrations is independent of the relative orientation of the field and the current. At high Ni concentrations the magnetoresistance is strongly orientation dependent. Based on the same arguments as discussed for the Ni-Ag system, we identify the isotropic negative magnetoresistance observed in samples with low Ni concentrations as a GMR-like effect, and the orientation-dependent magnetoresistance of samples with high Ni concentration as caused by an anisotropic magnetoresistance (AMR). Similar magnetoresistive behavior was found in another FI mixture, Co-SiO_2.

Prior to discussing the remarkable similarity between the two types of systems in the low ferromagnet concentration regime, we would like to address the topology of the systems under study. Due to the basic difficulties in microscopic analysis of crystalline metallic mixtures we do not possess an accurate topological map of metallic Ni-Ag and Co-Ag films. We, nevertheless, can conclude that the ferromagnetic material topology in these samples is similar to that in ferromagnet-insulator mixtures Ni-SiO_2 and Co-SiO_2, respectively. We are supported in this conclusion by (i) grain dimensions, as found in a transmission electron microscopy study; (ii) location of the percolation threshold; and (iii) magnetization measurements.

Grain size

Thin films with thicknesses of about 200 Å were deposited on graphite substrates and studied by transmission electron microscopy. Ni, Co, and Ag in Ni-Ag and Co-Ag films were found to be crystalline with typical grain sizes of the order of 30–40 Å. It is very difficult to distinguish individual grains in these metal-metal films. This prevents a credible definition of the spatial topology of the ferromagnet in a nonmagnetic metallic matrix. In Ni(Co)-SiO_2 films the ferromagnetic grains are crystalline, their size being about 30 Å. The grains are immersed in an amorphous SiO_2 matrix. The dimensions of the crystalline ferromagnetic grains are approximately equal in all the systems studied.

Percolation threshold

In metal-insulator mixtures the metallic percolation threshold can be identified by (a) the divergence of the resistivity and (b) the change of the resistivity temperature coefficient from positive in metallic regime above the threshold to negative in the dielectric regime below it. Based on these criteria, the concentration $x=49\%$ can be regarded as the percolation threshold of Ni in our Ni-SiO_2 mixtures.[16] A similar threshold was also found in Co-SiO_2.

In Ni-Ag the resistivity of both metallic components is very close and no singularity is found at the threshold. However, the magnetic properties of granular ferromagnets depend on the magnetic material concentration and, in particular, on the percolation of the magnetic phase. By crossing the magnetic percolation threshold a system such as Ni-SiO_2 starts exhibiting a bulk ferromagnetism[7] and associated with it anisotropic magnetoresistance. As we have reported previously,[16] the threshold in Ni-Ag was found at $x=47\%$. One can, therefore, conclude that both Ni-Ag and Ni-SiO_2 systems have the same ($\pm 5\%$) nickel continuity threshold.

Magnetization measurements

Additional information on the topology of the systems can be extracted from the magnetization measurements. The latter were performed in a superconducting quantum interference device magnetometer at room temperature at fields up to 2 T. A comprehensive discussion of the magnetization studies is beyond the scope of this paper and we shall demonstrate here only a few characteristic results. We plot in Figs. 3(a) and 3(b) the magnetization of two series of Co-based samples: Co-Ag and Co-SiO_2 as a function of an applied magnetic field. The range of Co concentrations is $37 < x < 62\ \%$ in both series. As one sees, the magnetization behavior is almost identical both quantitatively and qualitatively for all the concentration range. Since magnetic properties of granular ferromagnets depend mainly on their topology, we can conclude that the geometric structure of ferromagnetic material in metallic Ni-Ag and Co-Ag films is similar to that in Ni-SiO_2 and Co-SiO_2, respectively.

We turn now to the discussion of the GMR-like phenomena in granular ferromagnets. We define the normalized GMR effect as $(\rho_0-\rho)/\rho_0$, where ρ_0 and ρ are resistivities measured at zero and 10 kOe field, respectively. The values of the effect measured in all four systems: Ni-Ag, Ni-SiO_2, Co-Ag, and Co-SiO_2 are plotted in Fig. 4 as a function of ferromagnetic (Ni, Co) volume fraction. Two major conclusions can be drawn. (i) The largest normalized GMR effect occurs below the percolation threshold. In both FM and FI mixtures the maximum normalized GMR requires about the same concentration: 45–47 % for Ni-based mixtures and 40–

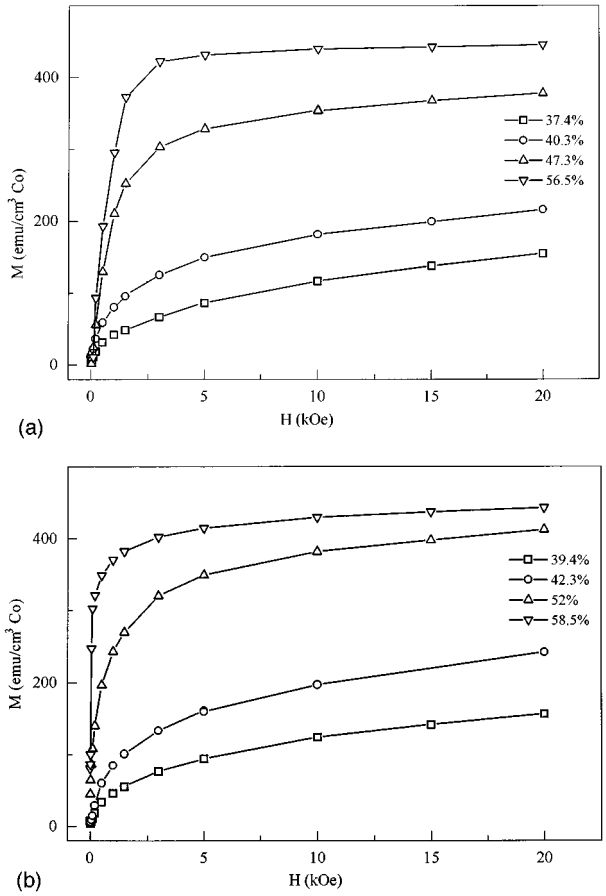

FIG. 3. Room temperature magnetization of Co-Ag (a) and Co-SiO_2 (b) samples as a function of magnetic field for a few different Co concentrations. The samples are 450-nm thick.

42 % for Co based mixtures. (ii) Despite the huge difference in the resistivities of FM and FI systems, the magnitude of the GMR effect is nearly the same in FM and FI mixtures. The maximum GMR is about 0.6% both in Ni-Ag and Ni-SiO_2. In Co-based films the maximum magnetoresistance was found to be an order of magnitude higher than in Ni-based compounds and reached about 10% in Co-Ag and

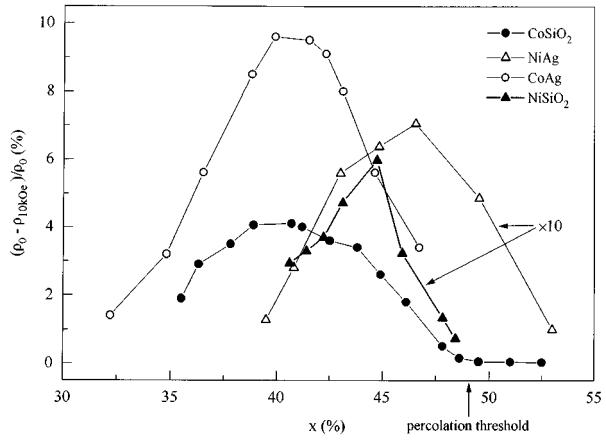

FIG. 4. Normalized GMR magnetoresistance as a function of the ferromagnet material component concentration x. The results for Ni-based series are multiplied by 10.

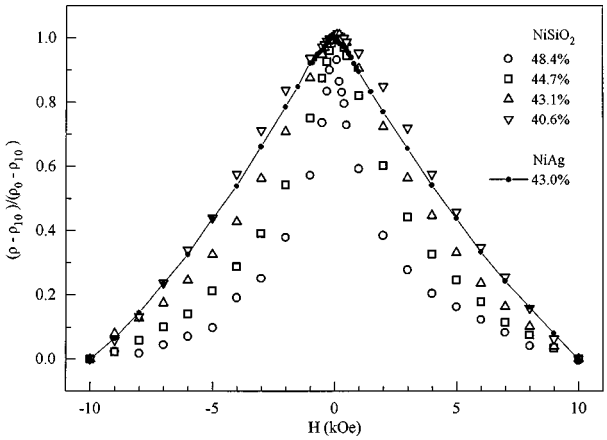

FIG. 5. Magnetoresistance of Ni-SiO_2 and Ni-Ag granular ferromagnets for different Ni volume concentrations normalized at zero and 10 kOe field. Put attention on the variation of the curves' shape.

4.5% in Co-SiO_2. The difference between the Co-SiO_2 and Co-Ag series is negligible compared to the ratio of the absolute resistivities 10^6:1 at $x=38\%$ and 10^7:1 at $x=32\%$.

A note should be taken here. The value of the magnetoresistance is defined in this paper as $\Delta\rho=\rho_0-\rho(10\text{ kOe})$, where ρ_0 and $\rho(10\text{ kOe})$ are resistivities at zero and 10 kOe fields, respectively. Actually, none of the samples studied showed the saturation of its resistivity at field of 10 kOe. We present in Fig. 5 the resistivity versus field curves normalized at zero and 1 T fields for several Ni-SiO_2 and Ni-Ag samples with different Ni concentrations. The slope $d\rho/dH$ gradually becomes sharper near the field of 1 T for lower Ni concentrations. This suggests that the saturation field increases for lower Ni concentration and the magnetoresistance measured up to the 1 T field is a fraction of the total saturated magnetoresistance which was not reached in these experiments. As reported by Ref. 19 magnetic fields higher than 14 T are required to reach magnetoresistance saturation in Fe-Ag films at low iron concentrations. High-field experiments are currently under way and will be reported elsewhere. However, as shown in Fig. 5, magnetoresistance curves of Ni-SiO_2 and Ni-Ag are very similar, and although we discuss a partial effect only, the comparison between the two types of systems is credible.

Let us compare in more details the dependence of the magnetoresistance on the ferromagnet material concentration x in ferromagnet-normal-metal FM versus ferromagnet-insulator FI mixtures. The GMR seems to develop in $NiSiO_2$ systems much sharper than in NiAg (see Fig. 4). Indeed, $\Delta\rho/\rho_0$ reaches its maximum of about 0.6% in both systems in the vicinity of $x=45\%$. No GMR behavior is observed in $NiSiO_2$ at concentrations above 48%, whereas the GMR-like magnetoresistance of the order of 0.2% is observed in NiAg at x as high as 53%. The percolation threshold of Ni in both $NiSiO_2$ and NiAg is about 48%. At concentrations higher than x_c Ni builds up both an infinite cluster and finite clusters of different sizes. Since the electrical current in NiAg system flows roughly uniformly in an entire volume, it tests both finite clusters and an infinite cluster simultaneously. The ferromagnetic phase percolates along the infinite cluster and along large enough finite clusters, giving rise to the aniso-

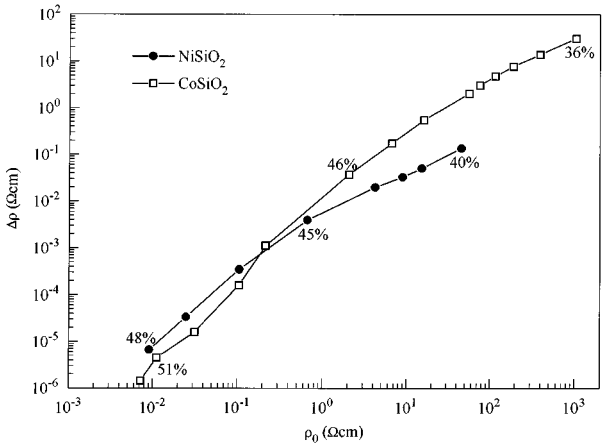

FIG. 6. Absolute value of the GMR-like magnetoresistance as a function of the zero field resistivity for Ni-SiO_2 (circles) and Co-SiO_2 (squares) samples.

tropic magnetoresistance. Small separated clusters can remain superparamagnetic and are responsible for the partial GMR contribution. Coexistence of AMR and GMR in the same samples above the percolation threshold is well illustrated by the ''winglike'' magnetoresistance curves in Fig. 1.

The situation is different in $NiSiO_2$ system. Here, from the moment when the infinite metallic cluster is created, all the current will flow along it and no current will pass between small grains separated by insulating gaps. Therefore, no GMR-like contribution is observed above the percolation threshold in FI sample. The anisotropic magnetoresistance effect is also negligible in the FI sample in the vicinity of the percolation threshold. We can mention at least two reasons for that. (i) AMR is a property of a bulk ferromagnet, in our case pure Ni. Due to strong scattering on the boundaries of the conducting paths, the resistivity of $NiSiO_2$ in the vicinity of the percolation threshold is much higher than that of pure Ni, and any variation of the bulk nickel resistivity is negligible on the total background. (ii) Contrary to the all-metallic systems, the local current direction in the branched and complicated topology of the FI samples can be oriented at any angle with respect to the local magnetization. Therefore, AMR becomes visible in $NiSiO_2$ at much higher Ni concentrations above 80% when the total resistivity decreases to about 10^{-4} Ω cm and the current flow becomes uniform.

Although the relative GMR-like magnetoresistance appears identical in ferromagnet-normal metal and ferromagnet-insulator mixtures, the absolute values of magnetoresistance demonstrate different tendency at low concentrations of ferromagnetic materials. In ferromagnet-normal metal mixtures Ni-Ag and Co-Ag the zero field resistivity remains roughly constant in an entire range of the composite concentrations. Therefore, both the absolute and normalized magnetoresistance show the same dependence on the ferromagnetic concentration. Namely, it develops in the vicinity of the percolation threshold, reaches maximum and decreases at low Ni/Co concentrations (see Fig. 4). The behavior is different in ferromagnet-insulator mixtures. We plot in Fig. 6 the absolute magnetoresistance value $\Delta\rho$ as a function of the room temperature resistivity ρ for Ni-SiO_2 samples. The absolute magnetoresistance value of Ni(Co)-SiO_2 mixtures does not show any tendency to decrease at low metal concentrations. Contrary to that, $\Delta\rho$ keeps increasing for the decreasing metal concentration, at least in the range we were able to measure.

Magnetoresistance of FM samples is often compared with their global magnetization. The connection between the magnetoresistance and global magnetization is understood to result from the sensitivity of GMR to the relative orientation of the neighboring magnetic moments. In the limit of the uncorrelated granular magnetic axes the average value $\langle\cos\theta_{ij}\rangle$, where θ_{ij} is the relative angle between the axes of the ferromagnetic entities can be described as: $\langle\cos\theta_{ij}\rangle=(M/M_s)^2$, where M is the global magnetization and M_s is its saturated value. Experimental agreement is reasonably good for certain samples, as shown in Fig. 7(a) for the Co-Ag sample with $x=43\%$.

The same comparison between the magnetoresistance and the global magnetization can be performed for Ni(Co)-SiO_2 films. Similar agreement is found in samples with low ferromagnet concentrations [Fig. 7(b)]. For higher ferromagnet concentrations in the vicinity of the percolation threshold the magnetoresistance stops following the square power of the global magnetization [Fig. 7(c)]. A simple explanation for this trend is topological. At concentrations close to the percolation threshold individual single domain grains coalesce into large ferromagnetic clusters. The latter dominate the global magnetization, whereas the GMR behavior is governed by the rest of uncoupled small superparamagnetic grains. Therefore, in this topological range, magnetization and magnetoresistance probe different parts of the sample: large ferromagnetic clusters and small superparamagnetic grains, respectively.

Both FM and FI systems seem to have a similar dependence of the magnetoresistance on the total magnetization and, therefore, similar sensitivity to the relative orientation of the neighboring magnetic moments. However, the roughly equal values of the ratio $\Delta\rho/\rho$ in both metallic and dielectric types of the systems is not *a priori* expected and remains to be understood.

In metallic FM systems GMR is traditionally regarded as the *extra* electrical resistance due to scattering from nonaligned ferromagnetic entities. At finite temperatures, the total resistivity of a granular metallic ferromagnet is usually described[20] as

$$\rho(H,T)=\rho_0+\rho_{\rm ph}(T)+\rho_m(H,T), \tag{2}$$

where ρ_0 is the temperature independent contribution from defects, $\rho_{\rm ph}(T)$ is the phonon contribution, and $\rho_m(H,T)$ is the magnetic resistivity responsible for GMR. The fractional GMR is then given by

$$\frac{\Delta\rho}{\rho}=\frac{\rho_m(0,T)-\rho_m(H,T)}{\rho_0+\rho_{\rm ph}(T)+\rho_m(0,T)}. \tag{3}$$

Each type of scattering is assumed to be independent and it's hard to imagine that the change of the total resistivity by many orders of magnitude will cause the same change in the spin-dependent contribution.

As we have mentioned above, the isotropic negative magnetoresistance was found by Gittleman *et al.*[17] in discontinuous Ni films more than two decades ago. The effect was treated by Helman and Abeles[8] in the framework of the tun-

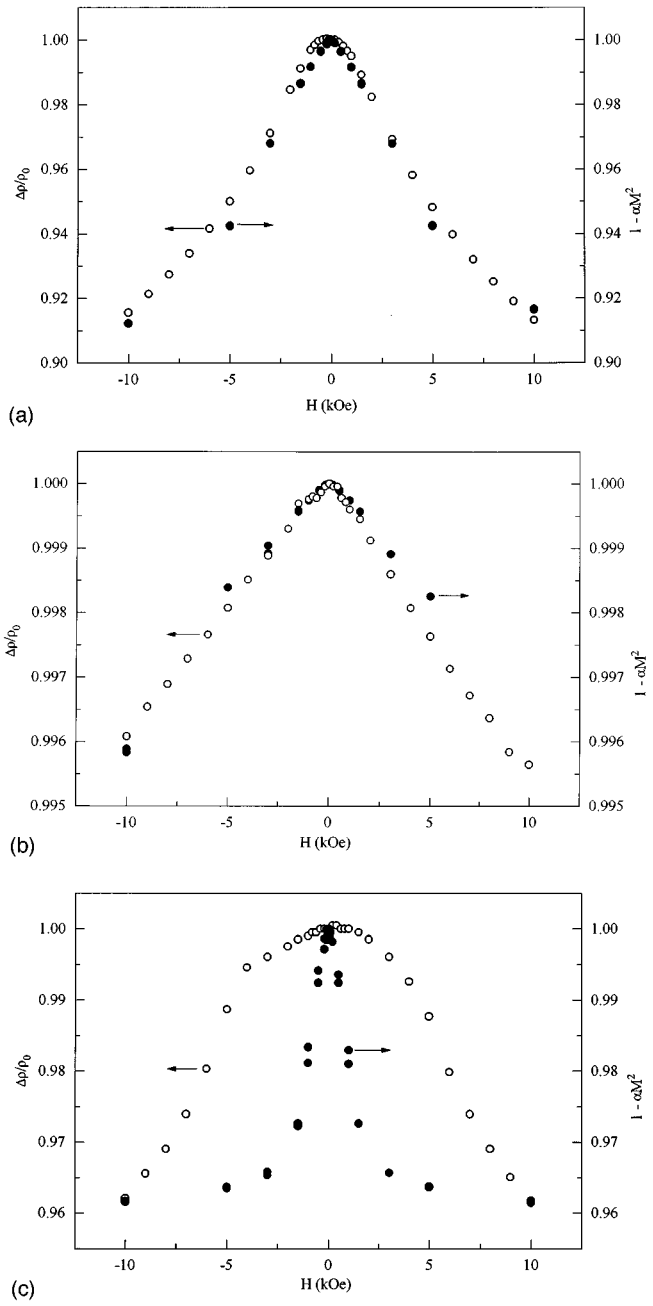

FIG. 7. Normalized magnetoresistance (open circles) and magnetization square M^2 (full circles) as a function of magnetic field for three typical samples. The data is normalized at zero and 10 kOe field. α is a normalization coefficient. (a) Co_xAg_{100-x}, $x=43\%$; (b) $Ni_x(SiO_2)_{100-x}$, $x=42\%$; (c) Co_xAg_{100-x}, $x=46\%$.

neling theory of spin-polarized electrons. Here, the effective intergranular barrier energy is represented by the sum of two contributions, electrostatic and magnetic. The electrostatic barrier energy includes the charging energy term, which becomes particularly important for small grains. The magnetic term E_m is the difference between the exchange energies of an electron situated in grains 2 and 1, which arises when the magnetic moments of the grains are not parallel and electron spin is conserved in tunneling. The relative orientation of the neighboring magnetic moments is affected by the magnetic field, therefore, the variation of the magnetic exchange energy E_m is responsible for the magnetoresistance. In this

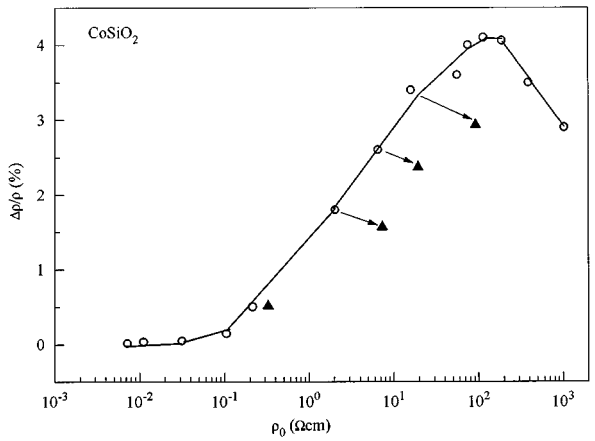

FIG. 8. Normalized magnetoresistance of Co-SiO_2 samples as a function of the zero field resistivity as measured after the deposition (open circles) and three months later (triangles).

model, the conductivity (σ) of the system is proportional to the probability of an electron tunneling across an intergranular barrier and is, therefore, proportional to the product of the nonmagnetic and the spin-dependent conductance terms.

$$\sigma(H,T)=\sigma_{el}(T)\sigma_m(H,T)=\sigma_0\exp(-E_m/kT). \quad (4)$$

For the superparamagnetic grains the magnetoresistance is given by[8]

$$\Delta\rho/\rho=-(JP/4kT)L^2(\mu H/kT), \quad (5)$$

where P is the polarization of the tunneling electrons, J is the electronic exchange coupling constant *within* the ferromagnetic metal grains, L is the Langevin function, and μ is the magnetic moment of the grain. Within this model the normalized magnetoresistance depends mainly on the intragranular properties, electronic polarization, the intragranular exchange coupling constant, the granular magnetic moment and does not depend on the total resistivity of the sample.

The independence of the effect on the resistivity of the ferromagnet-insulator sample is confirmed by the following data presented in Fig. 8. Here, the normalized magnetoresistance of a number of $CoSiO_2$ samples is shown as a function of their resistivity as measured shortly after the deposition and a few months later. Resistivity of $CoSiO_2$ samples increases with aging due to a partial dissociation of SiO_2 to SiO and O and the following oxidation of Co. The effective intergranular insulating layer becomes thicker which leads to an exponential increase of resistance. As seen in Fig. 8 the resistivity of the samples has increased up to about an order of magnitude, however, the normalized magnetoresistance almost did not change. A small decrease of the latter can be related to a development of the CoO layer on the Co grains surfaces and a respective reduction of the magnetic moments.

To conclude, we performed the first comparative study of magnetotransport phenomena in two types of granular ferromagnets: ferromagnet-normal metal mixtures Ni-Ag, Co-Ag and ferromagnet-insulator mixtures Ni-SiO_2 and Co-SiO_2. The geometric arrangement of the ferromagnetic material in metallic Ni-Ag and Co-Ag films is likely to be similar to that in Ni-SiO_2 and Co-SiO_2. The main experimental findings can

be briefly summarized as follows: (i) 3D granular ferromagnet-insulator mixtures demonstrate isotropic negative magnetoresistance similar to the GMR effect in heterogeneous ferromagnet-normal metal alloys; (ii) both in ferromagnet-metal and ferromagnet-insulator mixtures the largest normalized GMR-like magnetoresistance is found in samples below the ferromagnetic percolation threshold; (iii) despite the difference of many orders of magnitude in the absolute resistivities of ferromagnet-insulator and ferromagnet-metal mixtures, the values of the normalized negative magnetoresistance found in both types of the systems are close to each other; (iv) contrary to FM systems, the absolute magnitude of the magnetoresistance increases at low concentrations of the ferromagnet component in FI mixtures. Assuming that a striking similarity between the ferromagnet-normal metal and ferromagnet-insulating mixtures is not accidental, it is a theoretical challenge to provide a general description of the magnetotransport phenomena in both types of granular ferromagnets.

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