Giant magnetoresistance in hybrid magnetic nanostructures including both layers and clusters

P. Holody,* L. B. Steren, R. Morel, and A. Fert
Laboratoire de Physique des Solides, Université Paris-Sud, 91405 Orsay, France

R. Loloee and P. A. Schroeder
Department of Physics, Michigan State University and Center for Fundamental Material Research, East Lansing, Michigan 48824

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We report on giant magnetoresistance (GMR) effects in a type of magnetic nanostructure including both layers and clusters. Our structures are prepared by sputtering and include both continuous permalloy layers and cobalt layers broken into small clusters (islands). We obtain magnetoresistance ratios as large as 35% in less than 10 Oe, with slopes of about 6.5% per Oe in the steepest part of the curve. The advantage of such hybrid structures including layers and clusters, and the origin of the observed GMR effects are the following: (a) lateral decoupling of the thin Co layers into islands prevents ferromagnetic bridging by pinholes or other defects from extending to a significant part of the sample; (b) the field dependence of the GMR is controlled by the magnetization reversal in the thick and soft permalloy layers; (c) the antiparallel magnetic ordering is better as the contrast between the hard magnetic properties of the clusters and the soft ones of the layers is higher. The drawback of these hybrid systems is the crossover to superparamagnetic behavior as one goes to room temperature [clearly shown by superconducting quantum interference device (SQUID) measurements] and the resulting deterioration of the GMR slopes.

Giant magnetoresistance (GMR) has been discovered in magnetic multilayers[1,2] and then observed in a variety of magnetic nanostructures, multilayers, or cluster-based materials.[3] A large resistivity drop is obtained by aligning the moments of the magnetic entities, layers or clusters, by an applied field. Both types of systems—multilayered and cluster-based—are now in competition for low-field sensor applications. The drawback of the magnetic multilayers comes form the difficulty in preparing weakly antiferromagnetically coupled structures without additional parasitic coupling by pinholes, stray fields, etc. The general disadvantage of the cluster-based materials is that a relatively high field is necessary to overcome the anisotropy energy of particles with various shapes and align their magnetic moments.

Recently some attempts have been made to prepare intermediate systems between multilayers and cluster-based structures. Interesting results have been obtained by starting from multilayers and annealing them.[4–7] According to the interpretation of Bian *et al.*[6] or Hylton *et al.*,[7] by annealing, the nonmagnetic metal diffuses between the grains of the magnetic layers, so that these layers become discontinuous, that is, composed of disconnected islands or, in other words, pancake-shaped clusters. The advantage of such structures is well described in Ref. 7. The pancake-shaped clusters keep the low anisotropy of layers for in-plane fields. In addition, the structure with laterally decoupled magnetic islands is probably less sensitive to accidental bridging by pinholes or defects, since the lateral decoupling prevents such coupling from propagating at long distance. It has also been argued[7] that, for correlated arrangements of the islands in successive layers, dipole fields should favor antiparallel alignments of the island magnetic moments. Excellent MR results at low field have been obtained in annealed multilayers, for example a MR slope of about 0.8% per Oe, in NiFe/Ag.[7]

Another way to obtain intermediate structures between multilayers and cluster-based materials is to prepare multilayers with ultrathin and therefore discontinuous magnetic multilayers. This has already been done for Co/Ag with Co thickness of 4 and 6 Å.[8] Whereas, for thick Co layers, the probably existence of ferromagnetic bridging makes the MR very weak, a large MR begins to appear when the thickness of the Co is reduced and a MR ratio of 65% is reached for 4-Å-thick Co layers (with 15-Å-thick Ag layers). Large MR for Co/Ag with very thin Co layers has also been observed by Araki, Yasui, and Narumiya.[9]

In this paper, we report on the properties of a type of hybrid magnetic nanostructure including both thick permalloy layers and discontinuous ultrathin cobalt layers, for example, (Co 4 Å/Ag 35 Å/NiFe 20 Å/Ag 35 Å)$\times$15. Such structures combine the advantage of discontinuous Co layers (lateral decoupling) with the advantage of thick permalloy layers (high permeability and low coercivity). This leads to a high MR ratio at very low fields. For example, in some of our samples, at 4.2 K, we obtain a resistivity change of 34.5% in about 10 Oe and, at the steepest part of the MR curve, the slope amounts to 6.5% per Oe. However, for the samples we have prepared up to now, the Co clusters become superparamagnetic below room temperature (RT), which reduces considerably the MR. A better control of the cluster size is still needed to shift the superparamagnetic behavior above RT.

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Samples of the type (Co 4 Å/Ag t_{Ag}/NiFe t_{NiFe}/Ag t_{Ag})×15 were deposited by sputtering on Si(001) substrates, with values of t_{Ag} ranging from 10.5 to 40 Å, and with t_{NiFe} = 20 or 40 Å. We have chosen 4-Å-thick cobalt layers because we knew from our earlier work on Co/Ag that this thickness of Co leads to discontinuous layers and larger MR effects.[8] According to the NMR measurements of van Alphen, van der Heijden, and de Jonge[10] on samples with nominally 4-Å Co layers, the typical length scale of the "islands" is about 50 Å with an effective thickness around 10 Å. We have prepared series of samples with and without a 50-Å-thick Fe buffer layer on Si. Series have also been prepared at different substrate temperatures, ~−90, ~−60, and ~−25 °C. The results are nearly the same for samples with and without buffer. For samples prepared at different temperatures, we have observed only minor differences. Magnetization measurements with in-plane fields were performed with a superconducting quantum interference device magnetometer at temperatures between 5 and 300 K (some room-temperature measurements were also performed with a gradient magnetometer). Magnetoresistance was measured using a dc method between 4.2 K and room temperature.

In Fig. 1 we show magnetization versus field at 4.2 K for a sample with t_{Co}=4 Å, t_{NiFe}=40 Å, t_{Ag}=40 Å. The experimental curve is, without ambiguity, the superposition of a very sharp step (dashed-dotted line) coming from NiFe layers with very soft magnetic properties and of a broad loop (dashed line) corresponding to the discontinuous Co layers. Two important features can be noticed: (a) The Co remanent magnetization at low field is practically equal to its saturation magnetization, which means that the Co magnetization remains completely saturated in the small field range where the NiFe magnetization flips into the opposite direction. (b) The NiFe layers exhibit very soft magnetic properties with a coercive field of only 1.2 Oe. It thus turns out that the contrast between the hard and soft magnetic layers is more pronounced than in classical GMR systems such as Co/Cu/NiFe/Cu (our permalloy layers are softer and our cobalt layers harder).[11]

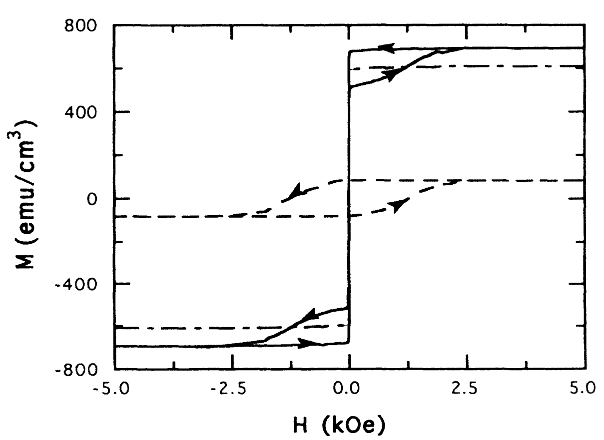

FIG. 1. Magnetization vs field at 4.2 K for a (Co 4 Å/Ag 40 Å/NiFe 40 Å/Ag 40 Å)×15 multilayer (with in-plane field). The total magnetization can be split between NiFe (sharp step, dashed-dotted line) and Co (large loop, dashed line).

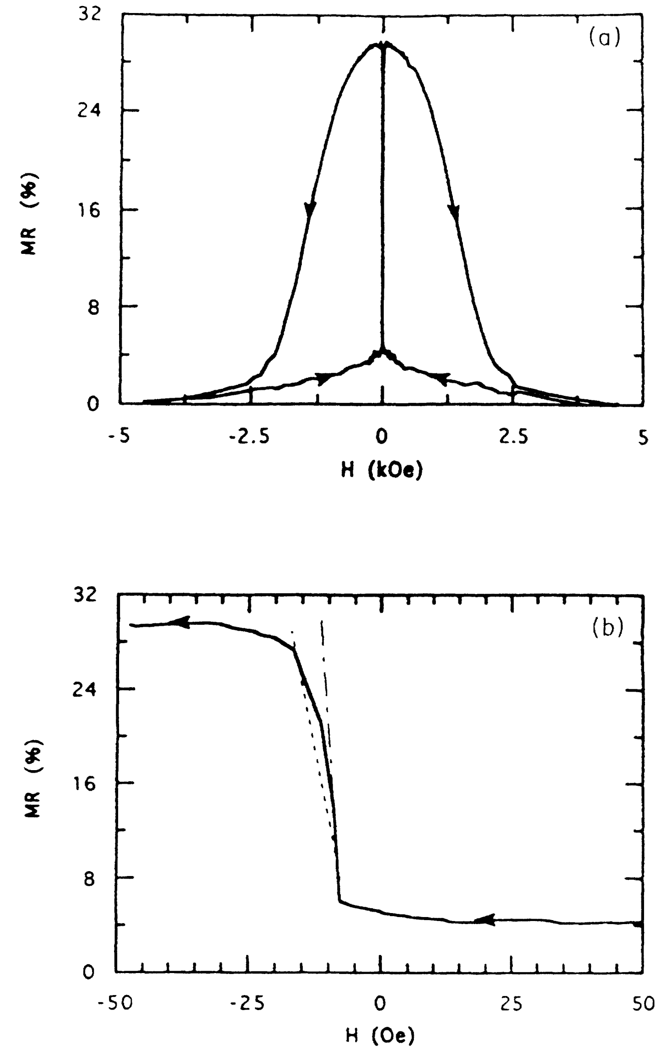

FIG. 2. (a) Magnetoresistance curves of a (Co 4 Å/Ag 40 Å/NiFe 40 Å/Ag 40 Å)×15 multilayer at 4.2 K in increasing and decreasing fields. (b) Blow up of the low-field part of the magnetoresistance curve in decreasing fields. The slopes of the two straight lines are 2.3% per Oe (dotted line) and 6.5% per Oe (dashed-dotted line), respectively.

The magnetoresistance curves of Fig. 2 confirm what would be expected from the magnetization curves of Fig. 1. The abrupt reversal of the NiFe magnetization gives rise to a steep increase of the resistivity in a field range of about 10 Oe. At somewhat higher fields, the resistivity remains almost constant and begins to decrease significantly only above 400 Oe, which is the field range (see Fig. 1) for the reversal of the cobalt magnetization. The total MR amounts to 30%. The blow up of Fig. 2(b) shows that a resistivity change of about 21.5% is obtained in a field range of only 9 Oe, which corresponds to a slope of 2.3% per Oe. The slope of the MR curve is not uniform and is steeper in the first 2 Oe: in this range the slope amounts to about 6.5% per Oe.

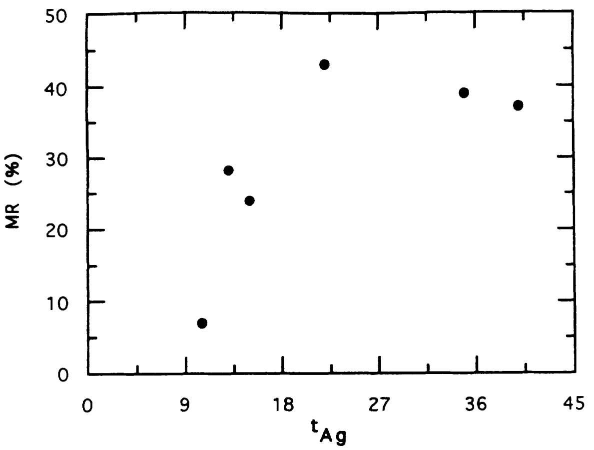

FIG. 3. Dependence of magnetoresistance on the thickness of Ag for (Co 4 Å/Ag 40 Å/NiFe 20 Å/Ag 40 Å)×15 multilayers deposited at about −45 °C on a 50-Å-thick Fe buffer layer (except for the sample with $t_{Ag}=40$ Å deposited at −60 °C without buffer).

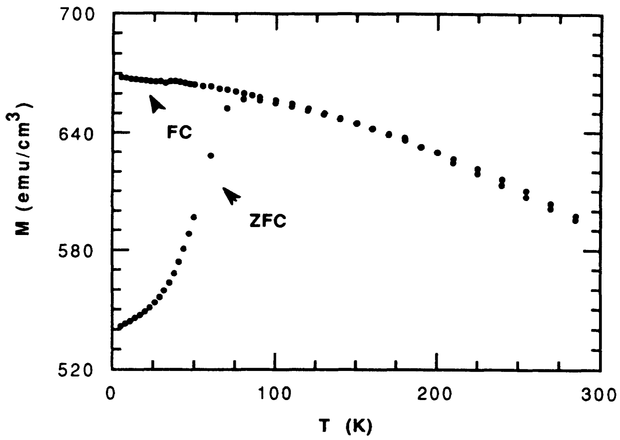

FIG. 4. Field-cooled (FC) and zero-field-cooled (ZFC) magnetization vs temperature for (Co 4 Å/Ag 40 Å/NiFe 40 Å/Ag 40 Å)×15. The applied field is 100 Oe.

We have obtained similar results for most samples. In Fig. 3 we show the variation of the MR ratio as a function of the thickness of the Ag layers. There may be a maximum at 13 Å, which could be due to a maximum of antiferromagnetic interlayer exchange in this range [this is in approximate agreement with the maximum at 15 Å observed in Co/Ag (Ref. 8)]. However, the MR ratio is only 28% at the first maximum, which shows that, for thin Ag layers, the MR is probably reduced by pinholes or other types of ferromagnetic bridging. The MR ratio reaches 43% for $t_{Ag}=22$ Å and then decreases slowly to 37% for 40-Å-thick Ag layers. If we consider now the slopes of the MR curves, it turns out that the softest NiFe layers are found for the thickest Ag layers. Our results are summarized in Table I. As already pointed out above, the results are practically the same with and without buffer layer, and also do not depend very much on the substrate temperature during deposition.

All the results presented above are for 4.2 K. At higher temperatures, the critical point is the crossover to a superparamagnetic behavior of the discontinuous Co layers. This crossover is clearly seen in Fig. 4 where the bifurcation between field-cooled FC and zero-field-cooled (ZFC) curves indicates the blocking temperature of the system of Co clusters. At the applied field of the measurements, the magnetization of the permalloy is saturated in FC and ZFC, so that the irreversibility seen at low temperature in Fig. 4 is due to Co layers. At room temperature (RT) the broad magnetization loop of the Co layer seen in Fig. 1 is no longer observed. The magnetization of the Co has become completely reversible.

The crossover to a superparamagnetic behavior of the Co clusters completely changes the GMR. Above the blocking temperature, the system of Co clusters has no more remanent magnetization and its magnetization is given by a Langevin law. At the very small field where the flip of the permalloy magnetization begins, cluster moments are randomly oriented, say half parallel, half antiparallel to the permalloy magnetization. Just after the permalloy flips, say 10 Oe after, the cluster moments are still randomly oriented, thus still half antiparallel, half parallel. Therefore the resistance does not change during the flip. The resistance will decrease significantly only at much higher fields needed to polarize the clusters. This is exactly what is observed in Fig. 5: the very

TABLE I. MR data for samples with similar thicknesses, $t_{Co}=4$ Å, 34 Å $\leq t_{Ag} \leq$ 40 Å, $t_{NiFe}=20$ or 40 Å) but deposited at different temperatures and with or without Fe buffer. We give the saturation value of the MR, the low-field MR with the field range in which this MR is observed, and the corresponding mean slope. As the MR slope is not uniform, the slope is generally higher when one considers only some part of the field range. For example, for the sample of the last line, a slope of 6.5% per Oe is observed in a field range of 2 Oe, see Fig. 2.

t_{Ag} Å)	t_{NiFe} (Å)	Deposition temperature (°C)	Fe buffer	Saturation Mr (%)	Low-field MR and corresponding field range	MR slope (% per Oe)
34	20	−3	No	38.8	37.5% in 20 Oe	1.9% per Oe
35	20	−41	Yes	38.9	23.6% in 15 Oe	1.6% per Oe
35	20	−62	No	39.5	34.5% in 10 Oe	3.5% per Oe
34	20	−93	No	35.7	26.5% in 15 Oe	1.8% per Oe
40	20	−62	No	37.2	35.1% in 10 Oe	3.5% per Oe
40	40	−62	No	29.7	21.5% in 9 Oe	2.3% per Oe

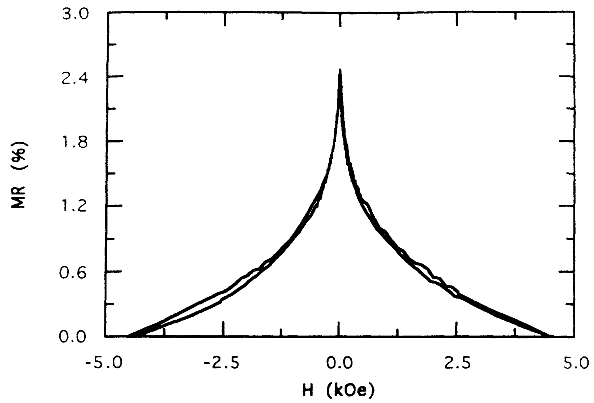

FIG. 5. Magnetoresistance curve of (Co 4 Å/Ag 40 Å/NiFe 40 Å/Ag 40 Å)×15 at room temperature.

abrupt resistance change at very small field seen in Fig. 2 does not exist at RT and we observe only the classical high-field MR of cluster-based systems. At 5 kOe, the MR is still not saturated. We have also recorded the MR curves at various temperatures. It turns out that the crossover from the behavior of Fig. 2 to that of Fig. 5 occurs in a relatively narrow temperature range, starting around 40 K below the blocking temperature seen in Fig. 4.

Extending the very interesting MR of Fig. 1 to room temperature for sensor applications requires an increase of the blocking temperatures to above 300 K. In principle, this can be obtained either by increasing the size of the Co clusters (enhancement of their anisotropy energy above kT) or by strengthening the ferromagnetic interaction between neighboring islands (at the possible cost of a decrease of the MR ratio). To increase the size of the Co clusters, we are now preparing new samples with slightly thicker Co layers and at slower deposition rates.

In conclusion, we have prepared hybrid magnetic nanostructures including both continuous magnetic layers (permalloy) and discontinuous magnetic layers composed of decoupled islands (cobalt). Our MR results at low temperature show a resistance change of as high as 35% over 10 Oe and steepest slope as high as 6.5% per Oe. These hybrid structures combine advantages of both multilayers and cluster-based materials: (a) The field dependence of the MR is controlled by the magnetization flip of soft permalloy layers, which explains why the MR is observed at very low fields; we point out that our NiFe layers are softer than those of Co/Cu/NiFe/Cu or NiFe/CuNiFe/FeMn structures,[11,12] which can be due to the weak miscibility of Ag in NiFe (compared to Cu) and also to the weakness of the exchange interaction across Ag (also the dipole fields generated by ultrathin Co layers are smaller). (b) Whereas Ag-Co is the best granular system for GMR, ferromagnetic bridging makes the GMR very hard to achieve in conventional Co/Ag or Co/Ag/NiFe multilayers. In our hybrid structure, the lateral decoupling between Co islands reduces the bridging effects and restores a large GMR close to that of the granular systems but at low field. Because our hybrid structures become superparamagnetic and lose their low field properties at room temperature, they are not yet useful for sensor applications. Our next step is to obtain high sensitivities at room temperatures by increasing the blocking temperatures above 300 K.

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*Permanent address: Department of Physics, Michigan State University, East Lansing, Michigan 48824.

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