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Giant magnetoresistance in soft ferromagnetic multilayers

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We show that the in-plane magnetoresistance of sandwiches of *uncoupled* ferromagnetic ($Ni_{81}Fe_{19}$, $Ni_{80}Co_{20}$, Ni) layers separated by ultrathin nonmagnetic metallic (Cu, Ag, Au) layers is strongly increased when the magnetizations of the two ferromagnetic layers are aligned antiparallel. Using NiFe layers, we report a relative change of resistance of 5.0% in 10 Oe at room temperature. The comparison between different ferromagnetic materials (alloys or pure elements) leads us to emphasize the role of bulk rather than interfacial spin-dependent scattering in these structures, in contrast to Fe/Cr multilayers.

The study of magnetic layered structures has resulted in the discovery of a variety of fascinating phenomena.[1] In particular, reports of giant magnetoresistance (GMR) in ferromagnetic multilayers (Fe/Cr) with antiferromagnetic coupling[2,3] have attracted great interest for fundamental physics as well as applications. More recently, similar effects have been found in other layered structures (Co/Ru, Co/Cr).[4] Enhanced magnetoresistive effects have also been found in Co/Au/Co sandwiches and multilayers with perpendicular anisotropy.[5–7] Various theoretical interpretations have been proposed for the observed magnetoresistive phenomena,[8–11] but fundamental understanding remains elusive.

In this paper we report very large magnetoresistive (MR) effects in sandwiches consisting of two *uncoupled* ferromagnetic layers with in-plane anisotropy separated by a nonmagnetic metal. This phenomenon, which we term spin-valve effect, is shown to be related to the relative angle between the magnetizations of the two ferromagnetic layers. It is different from the magnetic valve effect introduced by Slonczewski to describe the conductance perpendicular to the interfaces of sandwiches consisting of two ferromagnets separated by an insulating tunneling barrier.[10] The anomalous magnetoresistive effect is much larger than the usual anisotropic magnetoresistance (AMR) of these structures. We have observed it in several systems for which we were able to vary the relative orientations of the magnetizations of the different layers. The most demonstrative results were obtained from systems involving ferromagnetic pairs in which the magnetization of one member ($Ni_{81}Fe_{19}$, $Ni_{80}Co_{20}$, or Ni) was free to rotate while the other was constrained by exchange anisotropy through contact with antiferromagnetic $Fe_{50}Mn_{50}$. This exchange anisotropy is a unidirectional anisotropy which produces a shift of the hysteresis loop of the exchange-biased ferromagnetic layer[12] [see Fig. 1(a) for instance]. Using NiFe layers we have investigated the influence of several metallic interlayers. We have observed large effects with Cu, Ag, and Au while no effect was found with Al or Ta interlayers. The types of structures presented here extend the study of magnetoresistive effects to a broad, previously unexplored class of materials. Until the present study, investigations were limited to a small class of multilayers where the ferromagnetic layers exhibit antiparallel coupling or could be fabricated with different coercivities.[7] Our use of strong pinning created by exchange anisotropy makes possible magnetotransport studies of arbitrary ferromagnetic sandwiches exhibiting no interlayer coupling. These structures also permit a direct, quantitative measurement of the dependence of the resistivity on the relative orientation of the magnetizations,[13] each of which are forced to be in a single-domain state. Their very low saturation fields open the prospect for technological applications for sensitive field sensors.

The samples were prepared on Si substrates at ambient temperature in a computer-controlled dc magnetron sputtering system with a base pressure of 10^{-7} Torr. An argon plasma was used with deposition rates ranging from 0.5 to 2 Å/s. Resistance measurements were made with standard four point contact geometries with current in the film plane.

Figures 1(a) and 1(b) show, respectively, the magnetization curve and the change in resistance relative to parallel alignment, measured at room temperature (RT), for a sample with the following structure: Si/(150-Å NiFe)/(26-Å Cu)/(150-Å NiFe)/(100-Å FeMn)/(20-Å Ag). The magnetization curve shows two separate hysteresis loops. The loop with the smaller coercivity corresponds to the reversal of the bottom NiFe layer, while the loop shifted by exchange anisotropy to around 90 Oe corresponds to the reversal of the NiFe magnetization in the NiFe/FeMn bilayer. Thus, as the field H is swept, the magnetizations of the two NiFe layers change from parallel alignment for H lower than 2 Oe or higher than 135 Oe to antiparallel alignment between these two values. It is thus apparent that the change in resistance of Fig. 1(b) is related to the change in relative orientation between the magnetizations of the two ferromagnetic layers. For this sample the relative changes in resistance are 2% and 6%, respectively, at RT and 78 K. A 5.0% change in resistance at RT in 10 Oe was obtained for Si/(50-Å Ta)/(60-Å NiFe)/(20-Å Cu)/(45-Å NiFe)/(70-Å FeMn)/(50-Å Ta). This is comparable to the largest value observed for the AMR of bulk NiFe alloys but larger than any value reported for the AMR of thin films at room temperature.[14]

Using the structure Si/(60-Å NiFe)/(26-Å Cu)/(30-Å NiFe)/(60-Å FeMn)/(20-Å Ag) we have investigated the

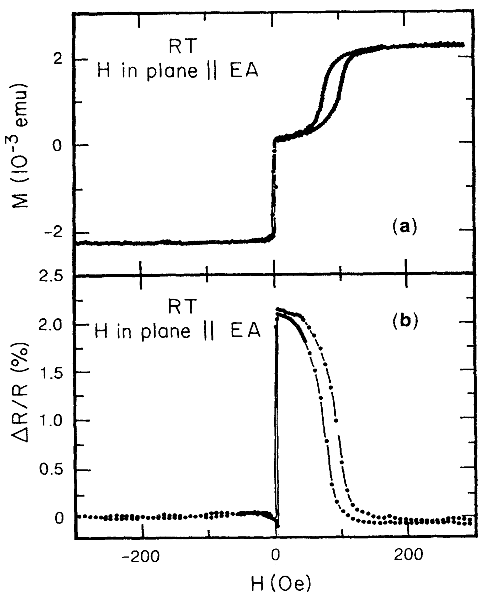

FIG. 1. Magnetization curve (a) and relative change in resistance (b) for Si/(150-Å NiFe)/(26-Å Cu)/(150-Å NiFe)/(100-Å FeMn)/(20-Å Ag). The field is applied parallel to the exchange anisotropy field created by FeMn (EA). The current is flowing perpendicular to this direction.

variation of magnetoresistance versus the angle $(\theta_1 - \theta_2)$ between the two magnetizations, see inset of Fig. 2. In this structure the NiFe/FeMn bilayer is exchange biased to 170 Oe, with its moment remaining nearly fixed in direction for fields up to $\simeq 15$ Oe, while the uncoupled NiFe layer can be saturated in any direction in the plane with fields larger than 7 Oe. Thus, by applying a 10 Oe rotating field one can rotate the magnetization of the soft layer without moving significantly the magnetization of the exchange-biased layer. Since θ_2 is nearly constant, to a good approximation $\cos(\theta_1 - \theta_2)$ is just the normalized component of the magnetization of the soft layer along the exchange anisotropy field H_{ex} (see inset Fig. 2). Two contributions are expected for the angular dependence of the magnetoresistance. The first one is the usual AMR, which is well-known to vary as the square of the cosine of the angle between the magnetization and the current. The second contribution is the spin-valve effect. We have directly measured the AMR on the same sample by comparing resistances for current applied parallel and perpendicular to the magnetizations. For both orientations we have used a field sufficiently high to saturate the magnetizations of the two NiFe layers. The AMR for only one layer was deduced using the relative thickness of the two layers. As shown in Fig. 2, we have subtracted this AMR contribution to single out the angular dependence of the spin-valve effect. Within our error bars, the angular dependence of the spin-valve effect is very well represented by a $\cos(\theta_1 - \theta_2)$ law. Quantitatively, the amplitude of the spin-valve effect is 3.05% compared to $0.37 \pm 0.02\%$ for the AMR of this structure. The latter value is smaller than for bulk NiFe partly due to shunting by the magnetically constrained Cu/NiFe/FeMn/Ag component of the structure and partly due to the increased resistivity of very thin NiFe layers.[14]

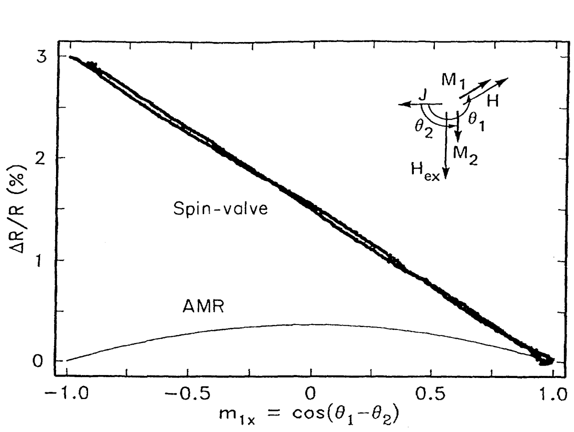

FIG. 2. Relative change in resistance vs the cosine of the relative angle between the magnetizations of the two NiFe layers of Si/(60-Å NiFe)/(26-Å Cu)/(30-Å NiFe)/(60-Å FeMn)/(20-Å Ag). Inset shows the orientation of the current J, exchange field H_{ex}, applied field H, and magnetizations M_1 and M_2.

We describe next the influence of the interlayer thickness on the magnetic and transport properties of films with structure Si/(50-Å NiFe)/(x Cu)/(30-Å NiFe)/(60-Å FeMn)/(20-Å Ag), with $x = 10$, 20, and 26 Å. The field is applied parallel to the exchange anisotropy field, the current is flowing perpendicular to this direction. As shown in Fig. 3(a) for $x = 10$ Å the two NiFe layers are

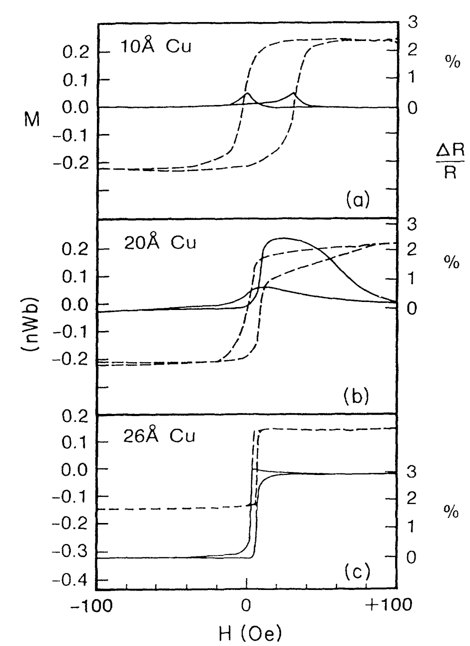

FIG. 3. Evolution of magnetization (dashed) and magnetoresistance (solid) curves for Si/(50-Å NiFe)/(x Cu)/(30-Å NiFe)/(60-Å FeMn)/(20-Å Ag) with Cu layer thickness $x = 10$, 20, and 26 Å. In (c), only the soft film reverses its magnetization direction in the field range ± 100 Oe.

strongly coupled, possibly due to the presence of pinholes or strong magnetostatic coupling in this range of thickness. As a result, the two magnetizations switch together without going through antiparallel alignment. The MR signal shows only two small peaks near the coercivity which may be ascribed to the AMR or scattering by domain walls.[12] We point out that an additional contribution to this peak may also come from local antiparallel alignment of the two layers during the reversal of the magnetization (see the discussion below). For $x=20$ Å, Fig. 3(b), the loops are nearly separated, but the switching process is not the same for increasing *versus* decreasing field. In the former case, the two magnetizations switch consecutively, while in the latter they switch together. Thus the MR signal displays a broad, rounded maximum due to antiparallel alignment in increasing field, but only a small peak for decreasing field. For $x=26$ Å, the two hysteresis loops are separated by more than 150 Oe, so that Fig. 3(c) shows only the switching of the unbiased layer. The magnetoresistance exhibits a 3% change over a very narrow range of applied field between 0 to 10 Oe, corresponding to the transition between parallel and antiparallel alignment. For samples with Cu thickness greater than 26 Å the amplitude of this change decreases due to increased shunting in the Cu layer and because diffuse scattering in the Cu layer reduces the flux of polarized electrons transmitted from one ferromagnetic layer to the other.

With NiCo substituted for NiFe the spin-valve effect is slightly smaller. For example, for Si/(50-Å NiCo)/(26-Å Cu)/(30-Å NiCo)/(70-Å FeMn)/(30-Å Cu) the spin-valve amplitude is 2.3% at RT instead of the 3% value seen with a comparable NiFe structure. Similarly, for structures of the same thicknesses using NiFe and noble metals the magnitude of the spin-valve effect decreases with the atomic weight of the interlayer material (1.25% $\Delta R/R$ for 100 Å of Cu, 0.7% with Ag, 0.5% with Au). Significantly, a fivefold decrease was seen for structures using Cu interlayers but with pure Ni substituted for NiFe or NiCo. Since Fe and Co impurities in Ni are known to have stronger spin-dependent scattering than Cu,[15,16] this suggests that the larger spin-valve effect seen in NiFe is dominated by the presence of 19% Fe in NiFe rather than to the presence of Cu impurities at the NiFe/Cu interface. Other experiments in which the NiFe thickness was varied confirmed this bulk rather than interfacial spin-dependent scattering mechanism.[17]

We stress that this is in contrast to theories of magnetoresistance in Fe/Cr multilayers where scattering at the interface is the suggested mechanism.[3,8,9]

We believe that the mechanism of magnetoresistance in the spin-valve effect is closely related to the giant MR effect previously observed in coupled systems such as Fe/Cr or Co/Ru multilayers. For these strongly coupled systems a two-current model with interfacial spin-dependent impurity scattering has been developed by several authors.[3,8,9] However, the results shown here lead us to emphasize the role of bulk versus interfacial scattering. The physical model of Ref. 3 can apply to our systems: In dilute NiFe alloys, it is known that the scattering of spin-down electrons on Fe impurities in the Ni host has a much higher cross section than the scattering of spin-up electrons[15,16] ($\alpha=\rho_{\downarrow}/\rho_{\uparrow}=20$). Thus for parallel alignment in the sandwich structure, the current is mostly carried by spin-up electrons which have long mean free path everywhere in the structure. The short circuit by the up electrons leads to the state of low resistance. On the contrary, for antiparallel alignment, both species of electrons are strongly scattered in the bulk of either one or the other ferromagnetic layer. The resistance is then increased compared to the previous case. Furthermore, we propose another contribution to the observed MR. Due to the nearly random trajectories of the electrons, each ferromagnetic layer emits and receives continuous flows of electrons. In the absence of current, these outgoing and incoming flows of electrons are balanced both with respect to the charge and spin. When a current flows through the sample, due to the spin-dependent scattering which occurs in the bulk of each ferromagnetic layer, the two species of electrons have different mean free paths. Consequently mainly the electrons with longer mean-free-path escape from the ferromagnetic layers, leading to a net polarization of emitted (and received) flows. For parallel alignment, a dynamical balance of spin is maintained between incoming and outgoing electrons. In contrast, for antiparallel alignment the two flows have opposite polarization so that an additional spin-flip relaxation mechanism must occur in the bulk of each ferromagnetic layer to maintain a steady state. This additional bulk spin-flip relaxation may also be part of the observed MR effect. If it is present, then there will be a change in the net magnetic moment of each ferromagnetic layer between parallel and antiparallel alignment. This contribution is expected to be much more important with current out of plane rather than in plane.

We believe the absence of the spin-valve effect with Al or Ta interlayers results from a decrease in the transmission of polarized electrons. The amplitude or the polarization of the flow or both may be affected. Interlayers of high resistivity (such as Ta), or interfaces of poor crystallographic quality may limit the exchange of electrons between the ferromagnetic layers.[18] Moreover it is possible that electrons are depolarized by paramagnetic Ni which may form along the interface.[19] It has been found that Al deposited on Ni (Ref. 20) and Ta deposited on NiFe (Ref. 21) create several monolayers of paramagnetic material. In addition, the spin-orbit interaction inside the interlayer may cause the depolarization of emitted electrons, consistent with the decrease of $\Delta R/R$ observed along the noble-metal series.[17]

The spin-valve effect may have been encountered but not recognized in other types of multilayers even if they did not consist of layers with unequal coercivity or of layers pinned by an antiferromagnet. In particular, sharp peaks in magnetoresistance near the coercivity have been observed in Co/Au/Co sandwiches[5,6] as well as in sputtered Fe/Ru multilayers.[22] These peaks may be partly attributed to local antiparallel alignment in neighboring layers due to uncorrelated nucleation of domains during the reversal of the magnetization. There may also be a fundamental correspondence between the spin-valve effect and three previously observed phenomena: the magnetic tunneling valve[10,11] seen for instance in Fe/Ge/Co,[23] the magnetoresistive effects observed in multi-domain single-crystal iron whiskers[24,25] and spin injection/detection.[26]

In summary, we have fabricated structures consisting of ferromagnetic layers separated by non-magnetic interlayers in which the change in resistance with the relative angle between the magnetizations is larger than anisotropic magnetoresistance. From the variation of the effect with different ferromagnetic as well as interlayer materials, we have identified scattering in the bulk of the ferromagnetic layers as the dominant mechanism for the spin-valve effect, in contrast to coupled systems such as Fe/Cr where scattering at interfaces is believed most important.

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