PHYSICAL REVIEW B **66**, 140404(R) (2002)

Spin filtering effect at inserted interfaces in perpendicular spin valves

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(Received 28 May 2002; published 29 October 2002)

Giant magnetoresistance can be enhanced by insertion of a nonmagnetic spacer in the ferromagnetic free layer of perpendicular spin valves. The enhancement and its independence of the inserted spacer thickness are argued in terms of the two-current series-resistor model, and interpreted in a simple picture that the *interfaces* between the inserted spacer and the parallel ferromagnetic layers act as spin filters. Magnetization of the laminated free layers behaves similarly to that of the single one; it indicates its applicability to the magnetic read head.

DOI: 10.1103/PhysRevB.66.140404 PACS number(s): 75.70.Pa, 72.25.Ba, 72.25.Mk, 75.70.Cn

There has been a lot of interest in devices that utilize the spin degrees of freedom of the charge carriers, the so-called ''spintronic'' devices.[1] Spin valves exhibiting a giant magnetoresistance (GMR) effect[2] are one of such devices, which have already been applied to the magnetic read heads for hard disk drives. However, the rapid growth of the recording density recently requires revolutionary improvement in the GMR properties.[3] Since it was revealed that the GMR effect in the multilayers in the current-perpendicular-to-plane (CPP) mode is larger than that in the current-in-plane mode,[4,5] the CPP GMR of the spin-valve multilayers is a topic of growing interest in GMR research.[6] According to the semiclassical theory,[7] the larger difference of the conductivity between spins up and down in the multilayers produces the larger CPP GMR. Applying highly spin-polarized materials such as half metals[8] to the GMR materials is an approach to obtain higher sensitivity, but it is difficult to prepare the practical GMR devices using those novel materials.[9] In this Communication, we show experimentally that inserted ferromagnetic-nonmagnetic (F/N) interfaces in a magnetic layer of the spin valves can act as spin filters and enhance the CPP GMR. The effect can be interpreted phenomenologically in the framework of the two-current series-resistor model.[7] It helps to comprehend the role of the interfaces in the spin valves in the CPP mode, and elucidate the physics of the CPP GMR.

Ordinary CPP spin valves consist of three metallic layers: a ferromagnetic free layer the magnetization of which rotates freely by an applied magnetic field, a nonmagnetic spacer, and a ferromagnetic pinned layer exchange-biased by an adjacent antiferromagnetic layer. The perpendicular current through the spin valve depends on the relative orientation of the magnetizations of the two magnetic layers, which can be controlled by the applied field, due to spin-dependent bulk and interface scattering.[7] In the structure we introduced,[10] one or more spacers are inserted in the free layer [Fig. 1(a)]. Here, as shown below, the magnetizations of the laminated free layers align ferromagnetically and behave as one ferromagnetic layer when a magnetic field is applied [Fig. 1(b)]. A simple picture of the effect based on the two-current model is as follows. Suppose resistivity of spin-down electrons is larger than that of the spin-up electrons in the ferromagnetic free layer in which a spacer is inserted. If spin-dependent scattering at the inserted F/N interfaces is sufficiently large, it enlarges total spin-down resistance of the laminated free layers without significant increase of the spin-up resistance of them. It means that the difference between the spin-up and spin-down resistance increases due to the additional F/N interfaces, i.e., the inserted interfaces act as spin filters in the layer. In other words, effective spin asymmetry of the system increases by the spin-dependent scattering at the inserted interfaces.

Our spin valves were sputter-deposited in the ultrahigh vacuum chamber and patterned by the photolithography and ion-milling processes. The element size varies from 0.1 μm^2 to 1.0 μm^2, which is comparable to the practical size for the magnetic read heads. The details of the fabrication process

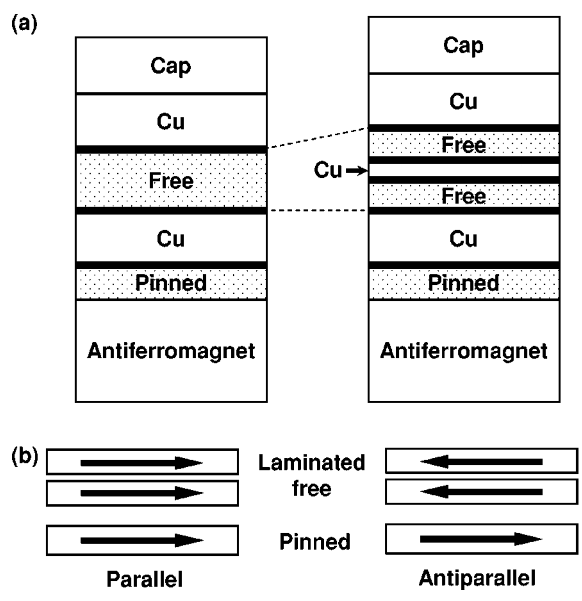

FIG. 1. (a) Schematic structure of the ordinal (left) and laminated free-layer (right) spin valves. Thick lines indicate ferromagnetic/nonmagnetic interfaces. (b) Direction of the magnetizations in the laminated free-layer spin valves when they are parallel and antiparallel.

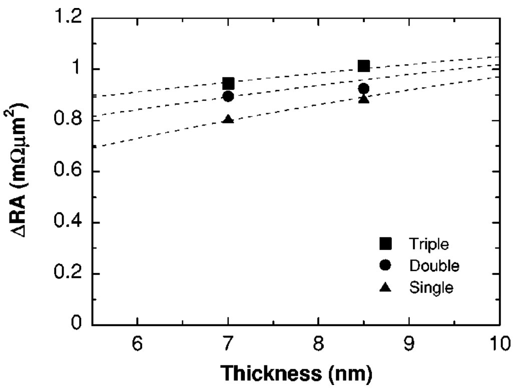

FIG. 2. Magnetic layer thickness dependence of ΔRA for single (triangle), double (circle), and triple (square) free-layer spin valves. Dashed lines are fits by the two-current series-resistor model [Eq. (1)].

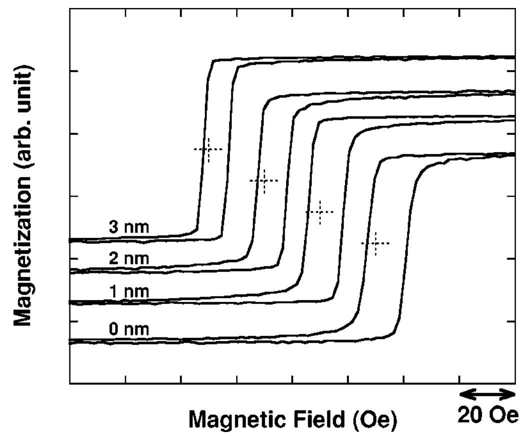

FIG. 3. Magnetization hysteresis loops for single and triple free-layer spin valves. Thickness of the inserted Cu spacers is 0 nm (a single free layer), 1 nm, 2 nm, and 3 nm. The curves are shifted both horizontally and vertically for clarity. The dotted crosses indicate their origins.

are described elsewhere.[11] We show here results for cases where one or two spacers were inserted in the free layer (hereafter we call them double and triple free-layer spin valves, respectively). The basic spin-valve structure is $Pd_{30}Pt_{18}Mn_{52}$ (PdPtMn) 13 nm/$Co_{88}Fe_{10}B_2$ (CoFeB) 2.5 nm/Cu 4 nm/free/Cu 4 nm/Ta 5 nm, where free = CoFeB, CoFeB/Cu/CoFeB, and CoFeB/Cu/CoFeB/Cu/CoFeB. To enable simple analysis, a single pinned layer was adopted for the GMR measurements. Similar enhancement, however, can also be observed when a commonly used synthetic ferrimagnetic pinned layer[12] is applied. All measurements shown below were made at room temperature.

In the CPP configuration, the intrinsic quantities are a product of the resistance (R) and magnetoresistance element area (A), and that of the resistance-change (ΔR) and A. We derive RA and ΔRA by plotting R and ΔR as a function of A measured by the scanning electron microscope. Several tens of the spin-valve elements were fabricated on a wafer varying the element size for each structure. R and ΔR for all the cases are almost inversely proportional to A, and RA and ΔRA are calculated by least-squares fits (data not shown). Figure 2 shows results for ΔRA as a function of the magnetic layer thickness (the sum of the free- and pinned layer thicknesses) for the single, double, and triple free-layer spin valves. Only the free-layer thickness is varied, and the Cu spacers of 1.5-nm thick are inserted at regular thickness intervals. The results clearly indicate increases of ΔRA up to 20% by the insertion of the F/N interfaces in the free layer. Note that each single point of the data in Fig. 2 is deduced from the experimental results for dozens of samples. Increments of ΔRA by thickening the magnetic layers is due to spin-dependent bulk scattering, while that by inserting the Cu spacers is due to additional interface scattering.

The magnetization measurements by an alternating-gradient magnetometer confirmed that the magnetizations of the laminated free layers rotate simultaneously with each other. Here the hysteresis loops of the triple free-layer spin valves were taken in the field range where only the magnetizations of the free layers are reversed. In order to make the pinned layer more stable to estimate the free-layer magnetization precisely, the synthetic ferrimagnetic pinned layer was used in these measurements. They show similar behavior and net magnetization within experimental errors irrespective of the spacer insertion and thickness as in Fig. 3. It is possibly due to the ferromagnetic interlayer coupling between the free layers separated by a thin spacer. This excludes the explanation that relative angle change between the magnetizations of the free layers contributes to the GMR increase. Figure 3 also shows that thickening of the inserted Cu spacers makes their coercive force slightly smaller. As the thinner CoFeB film is known to show the smaller coercive force, it is probably because ferromagnetic coupling between the free layers becomes weaker for thicker Cu spacers. The smaller coercive force will also be advantageous to the read head application.

We have modeled our results by applying a solution deduced from the Boltzmann transport equation[7] to our CPP geometry to expand on the qualitative picture mentioned above. Our model is similar to the two-current model used previously in Refs. 13 and 14 to describe regular CPP spin valves. In that model, the electrons are separated into two channels (spin up and down); they carry current independently. According to the theory, bulk and interface spin asymmetry coefficients β and γ are introduced; these parameters give the spin-dependent bulk resistance $\rho_{\uparrow(\downarrow)}=2\rho^*[1-(+)\beta]$ and interface resistance $R_{\uparrow(\downarrow)}A=2R^*A[1-(+)\gamma]$, respectively. Here ρ^* and R^*A can be described by the measured values ρ and RA as $\rho^*=\rho/(1-\beta^2)$ and $R^*A=RA/(1-\gamma^2)$. The in-plane resistivity was measured for CoFeB and Cu films fabricated in the same sputtering chamber, and the obtained values 25 $\mu\Omega$ cm and 4.2 $\mu\Omega$ cm from the thickness dependence were used in the calculation. For the F/N interface resistance, we have made micropillars that consist of CoFeB and Cu with different interface numbers, and deduced the value 1.0 mΩ μm^2. ΔRA of our system can be written as follows when the spin-diffusion length is much longer than each layer thickness and the diffuse scattering at the interfaces is ignored:

$$\Delta RA = \frac{4(2\gamma R^*An+\beta\rho^*t_F)(\gamma R^*A+\beta\rho^*t_P)}{(2R^*A+\rho_{\rm in}t_{\rm in})n+[R^*A-\rho_{\rm in}t_{\rm in}+\rho^*(t_F+t_P)+Z]}. \tag{1}$$

Here n is the number of the free layers, i.e., $n-1$ Cu spacers are inserted. t_F and t_P stand for the total thickness of the free layers, and the thickness of the pinned layer respectively. $\rho_{\rm in}$ and $t_{\rm in}$ are the resistivity and thickness of the inserted spacer. Z is the sum of the resistance for the CoFeB/PdPtMn interface, the Cu/Ta interface, and the rest of the Cu layers $[(R_{\rm CoFeB/PdPtMn}+R_{\rm Cu/Ta})A+\rho_{\rm Cu}t_{\rm Cu}]$; it is a free parameter for the fitting procedure here. At finite temperature the spin mixing resistivity should be taken into account for the precise calculation.[15,16] However, we assume it to be about an order smaller than the bulk resistivity for cobalt-based alloys even at room temperature,[17,18] and neglect the term for simplicity for our phenomenological analysis. It is also assumed that the spin-diffusion length for PdPtMn and Ta is so small that the resistance beyond CoFeB/PdPtMn and Cu/Ta interfaces can be excluded from the denominator of the ΔRA calculation.[13,14]

In Fig. 2, calculated ΔRA is also shown for the single and laminated free-layer spin valves by the dashed lines. From the fit to the experimental results, $\beta=0.58\pm0.05$, $\gamma=0.34\pm0.01$, and $Z=2.0\pm1.2$ mΩ μm^2, are obtained. Roughly, $\beta\rho^*$ decides the slope of the fitting curves, while γR^*A determines the n dependence. Z shifts the whole curves without significant change in the slopes and intervals, but is not so influential. Note that we have refined the parameter values used in the calculations by the subsequent experiments, and hence the calculated parameters β and γ are also varied from those obtained in the preliminary fits.[10] Systematic increases of ΔRA by the increment of the F/N interfaces in the magnetic layer can be well reproduced by the model as shown in Fig. 2. The point is that the simple picture based on the two-current series-resistor model is fairly valid for the submicron-sized spin valves consisting of the several ferromagnetic layers. The bulk and interface contribution to the GMR is derived, which can be a guideline for the spin-valve design.

It is important to understand the condition for ΔRA to increase by the spacer insertion. From Eq. (1), ΔRA is a monotonically increasing function of n when the relation below holds:

$$\frac{\gamma R^*A}{\beta\rho^*t_F} > \frac{2R^*A+\rho_{\rm in}t_{\rm in}}{2[R^*A+\rho^*(t_F+t_P)-\rho_{\rm in}t_{\rm in}+Z]}. \tag{2}$$

The relation can be interpreted in such a way that the inserted interfaces increase ΔRA if the spin asymmetry in the interface resistance (γR^*A) is large enough in comparison with that in the bulk resistance ($\beta\rho^*t_F$) to satisfy Eq. (2); our result discussed above is exactly the case. It should also be added that similar enhancement is expected from the theory when the spacer is inserted in the pinned layer. But practically, the ferromagnetic layers separated by the nonmagnetic spacer cannot be pinned strongly enough by a single antiferromagnetic layer. We, therefore, narrowed down the study to the insertion in the free layer here.

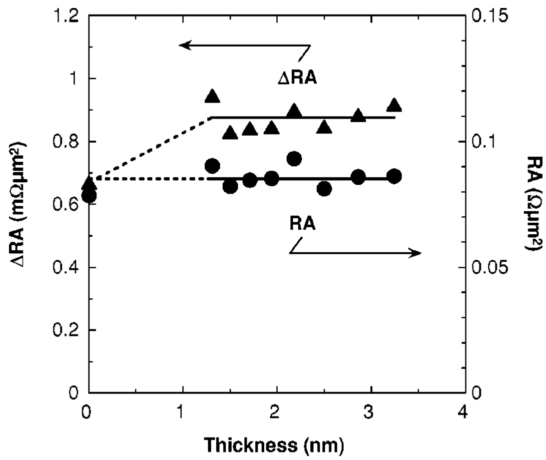

FIG. 4. RA and ΔRA as a function of the inserted Cu spacer thickness for double free-layer spin valves. Solid and dashed lines are guides for the eyes.

To elucidate that the enhancement is actually an interface effect, ΔRA and RA as a function of the inserted Cu-spacer thickness are shown in Fig. 4. When the thickness of the inserted Cu spacer of the double free-layer spin valves was varied from 1.3 nm to 3.2 nm, little change in ΔRA and RA was observed. Here the total free-layer thickness is 3 nm for all the samples. This independence coincides with what is expected from the theory. As the resistivity of the inserted Cu spacers is relatively small, RA and ΔRA are insensitive to the inserted spacer thickness when the distinct F/N interfaces are formed. Therefore, the fact strongly suggests that the observed increase in ΔRA is caused by the additional interfaces between the free layers and the inserted spacer, and that neither intermixing nor alloying of the materials occurs at least in the thickness range studied here. In addition, 3 nm of the Cu layer is thick enough to cover almost all of the layer, which indicates that pinholes do not play a role in the GMR enhancement.

From a technological point of view, we should comment on some merits of the laminated free-layer structure for the read head application. First, it shows currently 20% larger GMR with no sensitivity degradation only by adding a thin film of an already used material; no special extension is needed to fabricate this type of the spin valve. Furthermore, larger GMR enhancement can be expected by the optimal material combination and structure according to Eq. (1), which is currently under investigation. Secondly, the insertion can make the coercive force smaller with no change in the total free-layer magnetization; it leads to higher sensitivity. Thirdly, the total thickness scarcely increases by the insertion as opposed to, say, the dual-type spin valve; thin gap width is necessary for the read heads for high recording density.

In conclusion, we have demonstrated that the insertion of the Cu spacer(s) in the free layer of the spin valve can enhance the GMR effect. It is confirmed that the effect is due to the additional spin-dependent interface scattering, which can be regarded as the spin filtering effect at the inserted interfaces in the spin valve. The enhancement can be explained in the framework of the two-current series-resistor model. The laminated free-layer structure can give a clue to further understanding the CPP GMR, and also provide an additional method for future read heads.

The authors acknowledge S. Eguchi, C. Kamata, T. Fukuya, and H. Kubota for technical assistance for sample preparation. This work was partly supported by New Energy and Industrial Technology Development Organization (NEDO) under the Nanotechnology Program.

[1] G.A. Prinz, Science **282**, 1660 (1998).
[2] B. Dieny, J. Magn. Magn. Mater. **136**, 335 (1994).
[3] A. Tanaka, Y. Shimizu, Y. Seyama, K. Nagasaka, R. Kondo, H. Oshima, S. Eguchi, and H. Kanai, IEEE Trans. Magn. **38**, 84 (2002).
[4] W.P. Pratt, Jr., S.-F. Lee, J.M. Slaughter, R. Loloee, P.A. Schroeder, and J. Bass, Phys. Rev. Lett. **66**, 3060 (1991).
[5] S. Zhang and P.M. Levy, J. Appl. Phys. **69**, 4786 (1991).
[6] J. Bass and W.P. Pratt, Jr., J. Magn. Magn. Mater. **200**, 274 (1999).
[7] T. Valet and A. Fert, Phys. Rev. B **48**, 7099 (1993).
[8] Y. Ji, G.J. Strijkers, F.Y. Yang, C.L. Chien, J.M. Byers, A. Anguelouch, G. Xiao, and A. Gupta, Phys. Rev. Lett. **86**, 5585 (2001).
[9] W.E. Pickett and J.S. Moodera, Phys. Today **54** (5), 39 (2001).
[10] H. Oshima, K. Nagasaka, Y. Seyama, Y. Shimizu, S. Eguchi, and A. Tanaka, J. Appl. Phys. **91**, 8105 (2002).
[11] K. Nagasaka, Y. Seyama, L. Varga, Y. Shimizu, and A. Tanaka, J. Appl. Phys. **89**, 6943 (2001).
[12] J.L. Leal and M.H. Kryder, J. Appl. Phys. **83**, 3720 (1998).
[13] W.P. Pratt, Jr., S.D. Steenwyk, S.Y. Hsu, W.-C. Chiang, A.C. Schaefer, R. Loloee, and J. Bass, IEEE Trans. Magn. **33**, 3505 (1997).
[14] A.C. Reilly, W. Park, R. Slater, B. Ouaglal, R. Loloee, W.P. Pratt, Jr., and J. Bass, J. Magn. Magn. Mater. **195**, L269 (1999).
[15] A. Fert, J.L. Duvail, and T. Valet, Phys. Rev. B **52**, 6513 (1995).
[16] L. Piraux, S. Dubois, A. Fert, and L. Beliard, Eur. Phys. J. B **4**, 413 (1998).
[17] B. Loegel and F. Gautier, J. Phys. Chem. Solids **32**, 2723 (1971).
[18] J.L. Duvail, A. Fert, L.G. Pereira, and D.K. Lottis, J. Appl. Phys. **75**, 7070 (1994).