Enhanced robustness and tunnel magnetoresistance in artificial ferrimagnet based tunnel junctions

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Magnetic tunnel junctions have been fabricated, that use Co/Ru/Co and Co/Ru/CoFe artificial ferrimagnet (AFi) systems as hard magnetic electrodes. The ~22% tunnel magnetoresistance signal obtained, at room temperature, for junctions with Co/Ru/Co was increased to ~30% by taking advantage of the higher spin polarization of CoFe in contact with the barrier. Along with this improvement, a different reversal behavior for the AFi was obtained. Together with the enhanced thermal stability, up to 350 °C, of the junctions involving the Co/Ru/CoFe system potential use as magnetic sensors is discussed. © *2000 American Institute of Physics.* [S0021-8979(00)53108-9]

The discovery of large tunnel magnetoresistance (TMR) at room temperature in magnetic tunnel junctions (MTJs),[1] leads to the development of magnetic field sensors and memory applications. Basically, a MTJ consists of two ferromagnetic layers, separated by a thin insulating barrier. Since the resistance of the junction depends on the relative orientation of the magnetization of the ferromagnetic electrodes, with extreme values for the parallel and antiparallel alignment, the ferromagnetic layers must have different coercivities. For a given magnetic field the magnetically soft layer can reverse, while the magnetization of the hard layer remains unchanged. The amplitude of the resistance variation depends, among many other parameters, on the spin polarization values of the ferromagnetic layers in contact with the barrier. According to the simplified model proposed by Julliere,[2] the TMR, defined as the relative variation of the resistance between the parallel R_p and antiparallel state R_{ap}, is given by the relation: $\mathrm{TMR}=(R_{\mathrm{ap}}-R_p)/(R_p)=P_1P_2/(1-P_1P_2)$, where P_1 and P_2 are the spin polarization values of the ferromagnetic layers in contact with the barrier, meaning that the TMR signal is material dependent. In brief, a couple of hard and soft magnetic layers, with high spin polarization, are needed for industrial applications.

An emerging alternative for the hard magnetic system is the artificial ferrimagnet trilayer (AFi).[3] It consists of two ferromagnetic layers, antiferromagnetically coupled through a thin nonmagnetic spacer layer. The AFi system should behave as a magnetically rigid body in the operational field window. Using the AFi, the coercivity of the single magnetic layer, H_c^{SL} is, in principle, multiplied by a factor Q that is given by the relation: $Q=(m_1d_1+m_2d_2)/|m_1d_1-m_2d_2|$, where m_i and d_i are the magnetizations and thicknesses of the ferromagnetic layers that form the AFi. So, the following relation holds:

$$H_c^{\mathrm{AFi}}\simeq QH_c^{\mathrm{SL}}. \tag{1}$$

This paper focuses on the following aspects: (i) The control of the rigidity gain of the AFi by adjusting the magnetic thicknesses of the ferromagnetic layers; (ii) the role of interfaces and their thermal stability on the TMR signal and on the magnetic properties of the AFi; (iii) the increase in the amplitude of the TMR signal by choosing a ferromagnetic layer with high spin polarization in contact with the barrier.

All stacks have been fabricated using a high vacuum sputtering system, with a base pressure of 2×10^{-8} mbar. A Cr(1.6 nm)/Fe(6 nm)/Cu(30 nm) buffer layer[4] is sputtered on previously etched Si(111) substrates, followed by the deposition of the AFi trilayer, Co(1.8 nm)/Ru(0.8 nm)/Co(3 nm), or Co(2 nm)/Ru(0.8 nm)/$Co_{50}Fe_{50}$(3 nm). The tunnel barrier is formed on top of the AFi by rf Ar/O_2 plasma oxidation of a previously sputtered Al film. On top of the barrier, the magnetically soft CoFe(1 nm)/Fe(6 nm) detection bilayer is grown and the whole stack is capped with Cu(10 nm)/Cr(5 nm). Magnetic tunnel junctions are patterned by photolithography in areas of: $10\times10\,\mu\mathrm{m}^2$ up to $100\times100\,\mu\mathrm{m}^2$. The magnetic properties of the samples were studied using an alternating gradient field magnetometer (AGFM) and magnetic force microscopy (MFM). The magnetotransport measurements of the junctions were performed using a four-point technique, while the quality of the barrier was verified using XPS and TEM techniques.[4]

The coercivity of the individual ferromagnetic layer, H_c^{SL}, along with the Q value, determine the coercivity of the AFi system [Eq. (1)]. To study the coercivity of the single magnetic layer, pure Co(3 nm) layers capped either with Ru or Cu, and Co_xFe_{1-x}(3 nm) layers, of various atomic concentrations, capped with Ru, were sputtered on standard buffer stacks. The equiatomic composition for the alloy, $Co_{50}Fe_{50}$, was found to better combine high magnetic moment and coercivity. Figure 1 shows the MH loops for Co/Ru, Co/Cu, and $Co_{50}Fe_{50}$/Ru systems. The abrupt change of M at low fields (20 Oe) corresponds to the switching of the

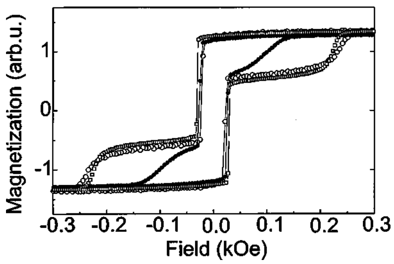

FIG. 1. Magnetization curves of a single Co(3 nm) layer capped with Ru (-●-) and Cu (-○-) and $Co_{50}Fe_{50}$(3 nm) layer capped with Ru (-□-).

Fe buffer layer and is not further discussed. These loops provide information about the magnetic/nonmagnetic metal interfaces in relation to the magnetic layer's reversal. Co is metallurgically miscible with Ru and this leads to interfacial mixing of the elements in the Co/Ru case. On the other hand there is no chemical affinity between Co and Cu, which results in very low intermixing.[5] The diffusion of Ru in the grain boundaries of the Co layer decreases the magnetic thickness of the Co layer and reduces the ferromagnetic coupling between the grains. The outcome is that the magnetization of Co(3 nm) capped with Ru is found to be about 1100 emu/cm^3, when the bulk value for Co is 1420 emu/cm^3 and its coercive field is lowered to about 100 Oe, compared to 220 Oe for Co/Cu (Fig. 1). Micromagnetic calculations also show that small thicknesses and weakly coupled grains favor reversal of the layer magnetization through rotation of the individual grain magnetic moments.[6] Indeed, the shape of the Co/Ru reversal (Fig. 1) suggests gradual rotation of the magnetizations in each grain. Limited diffusion at the interfaces with Ru is obtained with the use of $Co_{50}Fe_{50}$ alloy instead of Co. The high chemical affinity of Co and Fe prevents Ru from diffusing at the interface, hence keeping the magnetic moment of the alloy intact (~1800 emu/cc), while the coercive field, $H_c^{\mathrm{CoFe}} \simeq 220$ Oe, is similar to Co/Cu and the switching is much steeper than Co/Ru (Fig. 1).

The antiferromagnetic (AF) coupling, for both AFis, Co/Ru/Co and Co/Ru/CoFe, is found to have comparable strength. The oscillating behavior with the Ru thickness shows the first maximum at 0.3 nm and the second at 0.8 nm. The second AF maximum has been chosen in our AFi stacks in order to increase their thermal stability and avoid the presence of biquadratic coupling, which exists for the first AF maximum. In Fig. 2(a) the MH minor loops for the two systems of AFis are shown. The insets show the orientation of the magnetization inside the layers, with the first from the top and second arrow indicating the thick and thin AFi layer, respectively, while the third represents the Fe layer of the buffer. From the magnetic thicknesses of the layers, we calculate for the Co/Ru/Co $Q \simeq 4$, whereas for Co/Ru/$Co_{50}Fe_{50}$ $Q \simeq 2$. The coercive fields of Co/Ru/Co and Co/Ru/CoFe are about 400 Oe, in agreement with these expected from Eq. (1), considering the measured coercivity of the Co and CoFe single layer about 100 and 220 Oe, respectively (Fig. 1).

The difference in the reversal behavior of the two AFis stems from the different Q values and the quality of the interfaces. The Q factor relates to the stability of the magnetic domain walls. For strong AF coupled layers the walls appear in pairs and move as a unit. The pressure exerted on the walls is analog to the net magnetic moment of the AFi.[3] For a high Q, as in the case of Co/Ru/Co, the net magnetic moment of the hard system is low and the stability of the walls is enhanced, whereas for low Q, like in the case of Co/Ru/CoFe, the motion of the walls is facilitated by the large net magnetic moment. Moreover, in the stability of the walls for Co/Ru/Co, adds the fact that the nonmagnetic Ru defects, which have interdiffused into the Co at the interfaces, act as local wall pinning centers, as they reduce the magnetic surface of the walls and thus their energy. This effect can be seen in the MFM images for the two AFis [Fig. 2(c)], taken with an in-plane applied magnetic field of −600 Oe, after positive saturation. It is clear that, even though reversal of the AFi is almost completed, 360° domain walls still persist in the case of Co/Ru/Co, whereas for Co/Ru/CoFe walls have almost disappeared.[7] As a consequence of the above parameters, reversal of the Co/Ru/Co AFi takes place gradually, while the reversal of the Co/Ru/CoFe is driven more by the steep switching of the single CoFe layer and complies better to the picture of a single magnetic block.

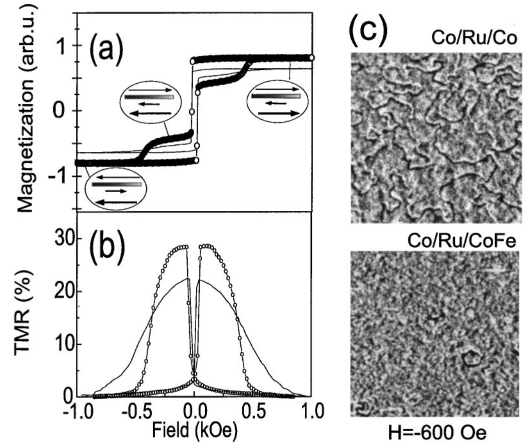

FIG. 2. (a) MH loops for buffer/Co/Ru/Co/Al_2O_3 (—) and buffer/Co/Ru/CoFe/Al_2O_3 (-○-) AFi systems, (b) MR loops for complete junction stacks that use Co/Ru/Co (—) and Co/Ru/CoFe (-○-) AFi, and (c) MFM images showing the stability of the walls for the Co/Ru/Co system and their disappearance for Co/Ru/CoFe.

The twofold larger coercive field of CoFe in comparison with Co allows us to use a smaller Q value to obtain similar coercive fields for the AFi. A way to enhance more the rigidity of the AFi, while keeping Q low, is by reducing the thickness of the alloy, and in this way increasing its coercive field.

The TMR signals for the junctions involving Co/Ru/Co and Co/Ru/CoFe AFi systems, as hard magnetic electrodes, are shown in Fig. 2(b). By using CoFe instead of Co as top AFi layer, a significant increase in the amplitude of the TMR value is succeeded, from about 22% at room temperature to about 30%. This improvement in the amplitude of the signal stems from the larger spin polarization of CoFe in comparison with Co, as expected from Julliere's model.[2]

The shape of the TMR curves reflects that of the MH loops of the AFis [Fig. 2(a)] and the presented MFM images

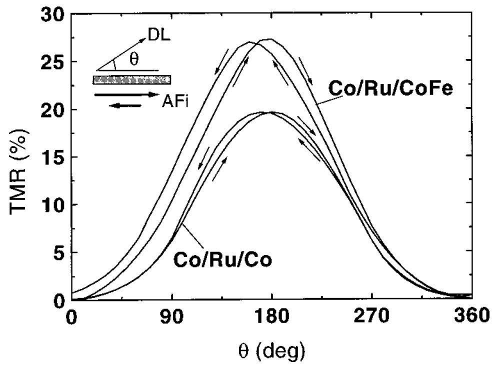

FIG. 3. Rotating field MR loops for junctions using Co/Ru/Co with $Q=4$ and Co/Ru/CoFe with $Q=2$. The applied magnetic field is $H=100$ Oe.

[Fig. 2(c)]. For the junctions that involve Co/Ru/Co, TMR drops very slowly, and sustains a relatively high value even after the reversal process has been almost completed. This relates to the stability of 360° walls, as discussed above, which act as high resistance channels for the tunneling electrons across the barrier.[7] On the other hand, the TMR signal for the junctions with Co/Ru/CoFe presents a steep drop, consistent with the rapid disappearance of the magnetic walls.

In Fig. 3 rotating field MR loops are shown, for both junction stacks and for magnetic field value of 100 Oe, beyond the coercive field of the detection layer. At this field, the net magnetization of the AFi is expected to be rigidly fixed and only the detection layer is free to rotate. Such a consideration would have led to nonhysteretic behavior. This is not the case, as can be seen in Fig. 3. The hysteresis for the junctions with Co/Ru/Co is reduced in comparison with Co/Ru/CoFe. This relates to the higher Q value of Co/Ru/Co than Co/Ru/CoFe and the consequent stability of the walls, as described above. Indeed, the larger Q value rigidly fixes the magnetic walls and prevents the domain structure to be influenced by the rotation of the external field.

A very important parameter for applications is the thermal stability of the junctions, which basically concerns: (i) The stability of the AFi system and (ii) the quality of the insulating barrier. To get insight on these two aspects the junctions involving Co/Ru/CoFe were subjected to vacuum anneal. As-deposited junctions show resistance-area products of about 80 $\mathrm{k}\Omega\times\mu\mathrm{m}^2$ and TMR values ranging from 27% to 30%. Parts of the wafer were annealed at temperatures from 150 to 350 °C. The TMR signal was slightly increased in the regime of 210 °C, by 1.5% (from 28.5% to 30%), after total annealing time of 2 h and 15 min, without any deformation of the shape of the signal, or change of the resistance (198 Ω for $20\times20\ \mu\mathrm{m}^2$ junction area). Successive annealing steps at 300 °C for 45, 60, and 80 min, affected neither the shape of the TMR curve nor its amplitude. The resistance slightly varied from 180 to 186 Ω for $20\times20\ \mu\mathrm{m}^2$ junction. A very interesting result is that, after annealing part of the wafer at 350 °C for 30 min, there was only a small decrease of the TMR signal, by about 1.5% and practically no change in the resistance (from 194 to 196 Ω for a $20\times20\ \mu\mathrm{m}^2$ junction), as shown in Fig. 4.

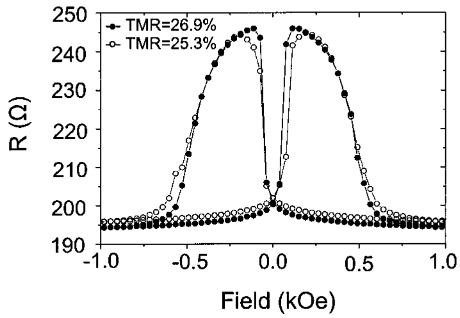

FIG. 4. TMR loops for the as-deposited (-●-) and annealed at 350 °C for 30 min (-○-) junction that uses Co/Ru/CoFe AFi system

The slight variations of the junction resistance for the whole temperature range suggest high stability and quality of the Al oxide barrier. Moreover, the almost constant amplitude of the TMR signal ascertains the stability of the interface between CoFe and Ru, which prevents intermixing even at high temperatures. This intermixing would cause structural degradation of the hard magnetic system, which would lead to its magnetic degeneration. The stability of both the resistance and magnetoresistance of the tunnel junctions, in contrast to other results in literature,[8] is extremely large up to 350 °C, making it attractive for applications because of the need of reduced resistances of the active systems, especially for the realization of magnetic heads.

In conclusion, this work aims at employing the large spin polarization of CoFe alloys for the fabrication of magnetic tunnel junctions that use the artificial ferrimagnet as hard magnetic electrode. Because of the larger coercivity of CoFe in comparison with Co, we can build Co/Ru/CoFe AFi with the same rigidity as Co/Ru/Co, but with smaller Q value. This leads to sharp reversal of the AFi and rapid disappearance of domain walls, which may be a significant advantage for magnetic sensors. The junctions using the Co/Ru/CoFe AFi show enhanced TMR signal of 30%, at room temperature, and thermal stability tested up to 350 °C. These characteristics make them good candidates for the realization of magnetic sensors and heads.

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