Enhancement of the thermal stability of magnetic tunnel junctions employing artificial ferrimagnets

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We have fabricated magnetic tunnel junctions that use Co/Ru/Co and Co/Ru/$Co_{50}Fe_{50}$ artificial ferrimagnet (AFi) systems as hard magnetic electrodes and AlO_x as tunnel barrier. The thermal behavior of the two AFis, incorporated in tunnel junctions, presents dramatic differences, the most remarkable being the much greater thermal stability of the Co/Ru/CoFe system. This stems from the improvement of the interfaces the CoFe alloy forms with its adjacent Ru and AlO_x layers. After successive annealing steps up to 400 °C, junctions incorporating the Co/Ru/CoFe system still present a significant tunnel magnetoresistance signal of nearly 20%, and most importantly, an almost intact magnetic rigidity of the hard magnetic system, being very promising for spin-electronic devices. © *2000 American Institute of Physics.* [S0003-6951(00)02048-9]

Following the discovery of large tunnel magnetoresistance (TMR) at room temperature,[1] magnetic tunnel junctions (MTJs) gained considerable interest, using state of the art thin film technology and being very promising for magnetic memories, head and sensor applications. MTJs basically consist of two ferromagnetic (FM) layers separated by a thin insulating barrier. The conduction of electrons through the barrier is spin dependent and, thus, modulated by the relative orientation of the magnetization of the two FM electrodes, with the resistance presenting extreme values for parallel and antiparallel alignment. For this, a pair of magnetically hard–soft electrodes is needed.

An alternative for the hard magnetic system is the artificial ferrimagnet (AFi),[2,3] consisting of two FM layers of unequal magnetic thicknesses, coupled antiferromagnetically (AFM) via a thin nonmagnetic layer. The coercivity amplification is quantified by the factor $Q=(m_1d_1+m_2d_2)/|m_1d_1-m_2d_2|$, where m_i and d_i are the magnetic moments and thicknesses of the FM metals forming the AFi. Details on the operation of the AFi can be found in Ref. 2. In this letter we will focus on the thermal stability of such a system, incorporated in a MTJ, which depends solely on the thermally activated diffusion processes at the interfaces between the magnetic layers, the nonmagnetic metallic spacer, and the insulating barrier. As a consequence, the quality of the interfaces which the FM metal forms with its adjacent layers, along with their chemical energy, are critical for the temperature stability of the MTJs. We show here that the use of CoFe instead of Co, in the AFi structure, can significantly improve the temperature response of the hard magnetic system and, thus, secure a high TMR signal at elevated temperatures.

We have prepared the MTJs by dc and rf magnetron sputtering on Si (111) wafers. On a standard Cr(1.6 nm)/Fe(6 nm)/Cu (30 nm) buffer,[3,4] the magnetic hard system has been sputtered, being either Co(1.8 nm)/Ru(0.8 nm)/Co(3 nm) ($Q=4$) or Co(1.8 nm)/Ru(0.8 nm)/$Co_{50}Fe_{50}$(2.8 nm) ($Q=2$). On top of the AFi the barrier was formed by rf Ar/O_2 plasma oxidation of a previously sputtered thin Al(1 nm) layer.[5] A magnetically soft bilayer of CoFe(1 nm)/Fe(6 nm) is used as a detection layer and the stacks are capped with Cu(5 nm)/Cr(3 nm). The junctions were patterned using UV photolithography in nominal areas of $10\times10\,\mu m^2$. The thicknesses of the AFi ferromagnetic layers have been chosen such that the switching fields of the two AFis would be similar—around 500 Oe [Fig. 1(c)]. The Ru thickness (0.8 nm) corresponds to the second AF maximum of the coupling oscillation curve, providing a high value ($J_{AF}=-2$ mJ/m^2) and small fluctuations of AF coupling with local changes in the Ru thickness. The Al oxide layer was shown to be of high quality in terms of low roughness, large scale continuity of the layer and proper oxidation conditions, as discussed extensively in Ref. 6.

The FM layers which form the hard subsystems are interfaced with both Ru and Al. Any intermixing with Al could be activated before the oxidation process takes place. To get an insight on the quality of interfaces, single layers of Co(3 nm) and CoFe(3 nm) were grown on the standard buffer (Cr/Fe/Cu) and capped with Ru(6 nm) and Al(3 nm). As a matter of reference, a Co(3 nm)/Cu(6 nm) sample was also prepared, since the interdiffusion between Co and Cu is known to be limited to only one single atomic layer.[7] The magnetization curves of these systems are shown in Fig. 1. The steep reversal at low field (20 Oe) is due to the seed Fe layer located in the buffer and will not be further discussed. The loss in coercivity of Co when interfaced with Ru [Fig. 1(a)] or Al [Fig. 1(b)] is clear as it drops to nearly half of the value obtained when interfaced with Cu, namely from about 220 to 100 Oe. Indeed, this softening of Co has to do with its high chemical miscibility with Ru and Al, leading to intermixed interfaces over three atomic layers, the first layer be-

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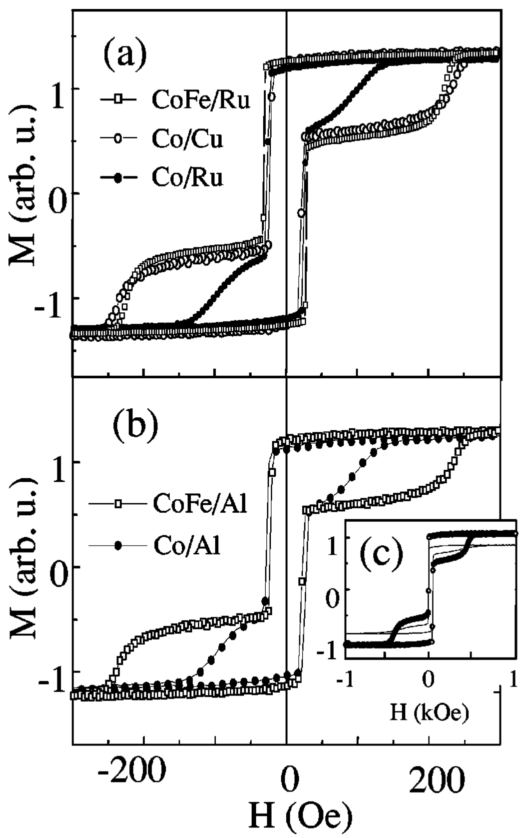

FIG. 1. (a) $M-H$ curves of single Co(3 nm) and $Co_{50}Fe_{50}$ (3 nm) layers capped with Ru (6 nm) and Cu(6 nm); (b) $M-H$ curves for Co(3 nm)/Al(3 nm) and $Co_{50}Fe_{50}$(3 nm)/Al(3 nm); (c) $M-H$ curves for the Co/Ru/Co (—) and Co/Ru/CoFe (-●-) AFi.

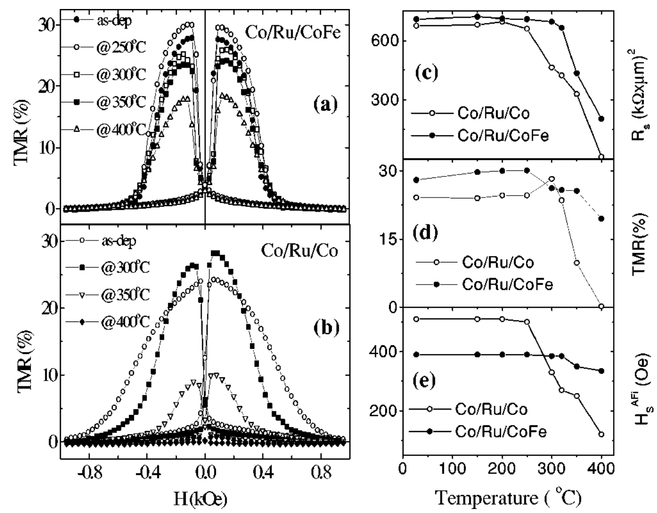

FIG. 2. Representative TMR loops for the as-deposited and postannealing cases for junctions employing (a) Co/Ru/CoFe and (b) Co/Ru/Co AFi. Temperature dependence of (c) the specific resistance, (d) of the TMR and (e) of the AFi switching field, for junctions incorporating Co/Ru/Co and Co/Ru/CoFe AFis.

ing magnetically dead.[7,8] The softening of the interfaces drags the bulk of the Co layer to switch at lower fields. The switching behavior becomes gradual and a loss in the magnetic moment appears, as shown in Figs. 1(a) and 1(b). In contrast, for the Co/Cu reference system, because of the limited intermixing, the switching is abrupt and the coercivity is much larger. Furthermore, the fact that the magnetization curves for CoFe/Ru(Al) and Co/Cu superimpose, gives strong indication that the mechanism of switching is very similar. Indeed, Co and Fe form extremely stable solid solutions for a large range of concentrations,[9] a fact which makes the alloy very reluctant to mix with Ru or Al at the CoFe/Ru(Al) interface. This interface stability gives rise to the large coercivity (220 Oe) and the steep switching of the CoFe/Ru(Al) systems, characteristics which denote the single magnetic body behavior of CoFe single layers. Because of the great interfacial quality the CoFe alloy offers, with all the consequential effects, its introduction in MTJs was primarily motivated to better respond in thermal treatment than Co.

The MTJs were subjected to successive annealing steps in the temperature range from 150 up to 400 °C for 60 min annealing time at each temperature. During annealing a nitrogen flow over the wafer protected the junctions from oxidation. In Figs. 2(a) and 2(b) the evolution of the TMR loop with temperature is shown for junctions employing Co/Ru/CoFe and the Co/Ru/Co AFi, respectively.

The evolution of the specific resistance with temperature gives information only on the thermal activation processes affecting the tunnel barrier, while the coercive fields and TMR variation with temperature reflects the thermal behavior of all layers in the stacks. In Fig. 2(c) we show the specific resistance dependence on the annealing temperature for MTJs containing Co/Ru/Co or Co/Ru/CoFe AFi. The specific resistance is rather stable at relatively low temperatures for both types of junctions. Then it steeply decays for temperatures above 250 °C down to two orders of magnitude at 400 °C for Co/Ru/Co [Fig. 2(c)]. The stability of the resistance is enhanced up to 320 °C for Co/Ru/CoFe with a rather limited effect at 400 °C [Fig. 2(c)]. The behavior of the barrier is mirrored on the temperature dependence of the TMR value, which, at 400 °C, drops down to zero for the Co/Ru/Co based junctions [Fig. 2(d)] while it decays by only 30% for the Co/Ru/CoFe based junctions [Fig. 2(d)]. This systematically observed behavior is attributed to the preferential degradation of the barrier when interfaced with Co, which presents a high affinity for Al. Given the appropriate thermal energy, Co atoms can locally be displaced towards the barrier, forming spin independent conductivity chains across the barrier and eventually lead to local breakdowns. The small increase of TMR in a specific temperature range is attributed to the oxygen kinetics in the FM metal/oxide interface, which improves the spin polarization.[10,11]

Beyond the temperature-dependent behavior of the AlO_x barrier, the other critical parameter for the TMR effect is the thermal stability of the hard and soft magnetic system. Figure 2(e) shows the temperature evolution of the coercivity for the Co/Ru/Co and Co/Ru/CoFe hard subsystems. The coercive field for the Co/Ru/Co AFi remains almost constant (~500 Oe) up to 250 °C, and then drastically decreases, reaching 120 Oe at 400 °C. This gradual decay of the magnetic rigidity stems from the thermally activated interdiffusion processes between Co and Ru, which further degenerate the hard subsystem structurally—and as a consequence—on its magnetic properties. The moderate thermal stability of the Co/Ru/Co AFi proves to be detrimental for the TMR effect at elevated temperatures [Fig. 2(d)], combined with the aforementioned phenomena related to the tunnel barrier. In-

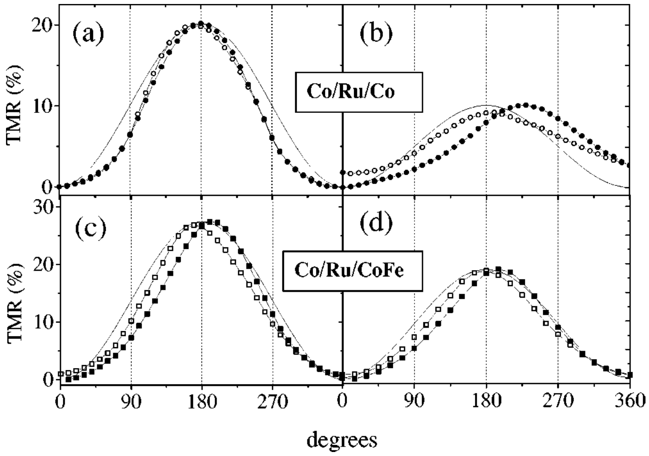

FIG. 3. Rotating field curves for junctions with Co/Ru/Co AFi: (a) ''as-deposited;'' (b) annealed up to 350 °C systems. Rotating field loops for junctions with Co/Ru/CoFe AFi; (c) as-deposited; (d) annealed up to 400 °C. The symbols on curves indicate: open circle (square) the ''forth'' curve, black circle (square) the ''back'' curve, no symbol (line): the θ fit curve.

deed, the TMR signal drops by a factor of 2 at 350 °C, related to a loss in the antiparallel alignment between the two FM layers in contact with the barrier. Although the coercive field is large enough to give rise to a significant TMR value at 400 °C, the complete loss of signal is mostly attributed to deterioration of the barrier as well as spin depolarization of electrons across the Co/AlO_x interface. In contrast, the coercive field of the Co/Ru/CoFe system remains constant at 390 Oe up to temperature of 320 °C and then experiences a slight decrease, reaching 340 Oe after successive annealing up to 400 °C [Fig. 2(e)]. This behavior is reflected on the temperature dependence of the TMR [Fig. 2(b)] even for extreme temperatures. Indeed, the high thermal stability of both CoFe interfaces with Ru and AlO_x allows a significant TMR value of nearly 20% after successive annealing up to 400 °C.

The physical processes taking place upon annealing are further illustrated by rotating field experiments realized for junctions incorporating either one of the two AFi subsystems. The applied rotating field value is 100 Oe, which should rotate only the soft-detection layer. Ideally, the variation of the TMR in a rotating field should follow a cosine function of the angle θ between the direction of the soft layer magnetization (coinciding with that of the applied field) and the fixed net magnetization of the AFi. The hysteretic features of the loops observed for the as-deposited cases [Figs. 3(a) and 3(c)] are due to the fact that the net moment in the hard subsystem is slightly deflecting towards the direction of the field. This is due to the random distribution of anisotropy axes within the grains constituting the FM layers of the AFi. The intralayer FM coupling between the grains and the interlayer AFM coupling between the AFi magnetic layers prevent large deflections. Co/Ru/CoFe based junctions present a larger hysteresis for the as-deposited state, than Co/Ru/Co based junctions [Figs. 3(a) and 3(c)], as the net moment of the AFi is larger in the first case ($Q=2$ for Co/Ru/CoFe and $Q=4$ for Co/Ru/Co), increasing the Zeeman energy term.

After annealing the Co/Ru/Co based junctions at extreme temperatures, the hysteresis increases [Fig. 3(b)], as the couplings are weakened due to the diffusion of Ru at the grain boundaries of Co. This favors the magnetic moment inside the grains to relax towards their local anisotropy axes, and rotate more or less individually when a rotating low magnetic field is applied. However, for the junctions using the Co/Ru/CoFe subsystem, even at 400 °C the magnetic behavior of the device remains basically unchanged compared to the as-deposited case [Figs. 3(c) and 3(d)]. It is a further confirmation of the rather limited diffusion within the CoFe layer. An interesting feature is that the bottom Co/Ru interface of the Co/Ru/CoFe AFi does not seem to affect the thermal properties of the AFi system, even though interdiffusion is expected. We must keep in mind that the grains of the bottom Co layer are strongly AF coupled with the grains of the top CoFe layer. So, even if Ru is diffusing into the Co and reducing the intralayer FM coupling, the strong coupling with the overlying CoFe layer would limit the misorientation of the Co grains.

In conclusion, we have fabricated Co/Ru/Co and Co/Ru/CoFe AFi based junctions. The introduction of the CoFe layer in the AFi enhances significantly the thermal rigidity of the junctions. This mainly stems from the improvement of the Ru/CoFe/AlO_x interfaces in terms of interdiffusion. Combined with the high spin polarization of the CoFe, a significant tunnel magnetoresistance signal of 20% can be achieved for junctions annealed up to 400 °C.

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