Large Magnetoresistance at Room Temperature in Ferromagnetic Thin Film Tunnel Junctions

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(Received 29 November 1994)

Ferromagnetic-insulator-ferromagnetic tunneling has been measured in CoFe/Al_2O_3/Co or NiFe junctions. At 295, 77, and 4.2 K the fractional change in junction resistance with magnetic field, $\Delta R/R$, is 11.8%, 20%, and 24%, respectively. The value at 4.2 K is consistent with Julliere's model based on the spin polarization of the conduction electrons of the magnetic films. $\Delta R/R$ changes little with a small voltage bias, whereas it decreases significantly at higher bias (>0.1 V), in qualitative agreement with Slonczewski's model. These junctions have potential use as low-power field sensors and memory elements.

PACS numbers: 75.70.–i, 73.50.Jt, 85.30.Mn, 85.70.Kh

Early experiments on spin-polarized tunneling between ferromagnets and superconductors [1] suggested the possibility of the magnetic-field dependence of tunneling between two ferromagnets separated by a thin insulator (FM/I/FM). In other words, the dependence of the tunneling current on the relative orientation of magnetization (M) of the ferromagnetic electrodes was expected. Using Co/Ge/Fe junctions, Julliere observed a change of nearly 14% at 4.2 K in the tunnel conductance at zero bias with the application of a magnetic field [2]. This effect became less than 1% when a few mV were applied to the junction. The fact that the barrier was 100 Å of amorphous Ge (a-Ge) and the absence of spin-polarized tunneling with a-Ge and most a-Si barriers found in later work [3] point toward a zero-bias anomaly in junctions with magnetic impurities [4], as Julliere has suggested. Later reports by various groups using mainly NiO and Al_2O_3 barriers between Ni and Co electrodes showed definite results for FM/I/FM tunneling [5–12]. However, in most of these cases, the change in the tunnel resistance $\Delta R/R$ was (2–7)% at 4.2 K, and only fractions of a percent at room temperature. Recent work by Miyazaki, Yaoi, and Ishio [7] showed a 2.7% change in the resistance at room temperature. In their experiment, part of the 150 Å Al film over the Permalloy film was oxidized to form NiFe/Al-Al_2O_3/Co tunnel junctions.

Julliere proposed an explanation of his result based on the conduction electron spin-polarization values of the FM electrodes, a model that later groups have essentially adopted [2]. According to this model, if P_1 and P_2 are the conduction electron spin polarization of the two FM electrodes, as measured by spin-polarized tunneling experiments with superconductors, the change in the tunnel resistance is given by

$$\Delta R/R = (R_a - R_p)/R_a = 2P_1P_2/(1 + P_1P_2). \quad (1)$$

Here R_p and R_a are the resistances with magnetizations of the electrodes parallel and antiparallel, respectively. For a Fe-Co tunnel junction, with P of 40% and 34%, respectively, for the two FM's, measured by the spin-polarized tunneling technique [1], the above expression gives a 24% change in the tunnel resistance between antiparallel and parallel orientations of M in the two FM electrodes. This is the ideal case, in the absence of limiting factors like domain walls in the junction area, interfacial and barrier spin scattering, direct coupling between the two FM films, and surface degradation of FM films, which could diminish the expected effect. Such factors certainly contributed to the low values of $\Delta R/R$ previously measured. Another theory of FM-FM tunneling has been proposed by Slonczewski [13]. This model analyzes the transmission of charge and spin currents through a rectangular barrier separating free-electron-like FM metals. It predicts strong influence of tunnel barrier height on the orientation of the spins tunneling across the FM-I interface, thereby affecting the spin polarization and also the exchange coupling between the ferromagnets, and was consistent with the low values of $\Delta R/R$ seen previously.

In the present work, some of the problems leading to low values of $\Delta R/R$ have been solved, significantly improving the results. Over a 10% change in the tunneling resistance with H has been observed consistently and is reproducible; in some cases as much as 11.8% change was seen. A sensitivity factor of about (0.13–0.2)%/Oe is obtained for the present tunnel junction devices. This increase in $\Delta R/R$ perhaps depends on a decrease in surface roughness, which can directly couple the two electrodes ferromagnetically, in addition to choosing good tunnel barriers.

FM/I/FM thin film planar tunnel junctions used in this study were prepared in a high vacuum (10^{-7} torr) evaporation system as follows. Initially the liquid-nitrogen-cooled glass substrate was covered with 10 Å of Si, followed by the first FM metal film, 80 Å thick and 0.2 mm wide. An aluminum film, 16–18 Å thick, was deposited to cover the entire first FM layer. After warming the substrate to room temperature, the Al film was oxidized by a glow discharge in oxygen at 60 μm to create the insulating tunnel barrier of Al_2O_3. (Spin-polarized tunneling with Al_2O_3 barriers has shown that this procedure oxidizes about

0031-9007/95/74(16)/3273(4)$06.00

12–14 Å of an Al film.) After pumping down again, cross strips of the top FM electrode 100–300 Å thick and about 0.3 mm wide were deposited at room temperature.

In each evaporation 72 junctions, with a junction area of about 6×10^{-4} cm^2, were prepared. Tunnel junction resistances ranged from hundreds of ohms to tens of kiloohms, depending on the duration of the glow discharge and the type of FM material used. Five junctions were selected from each set for resistance measurements as a function of H and T. The samples could be oriented with the film (junction) plane parallel or perpendicular to the field direction. Both ac lock-in and dc techniques were used for the resistance measurements. Tunnel conductance was measured as a function of applied bias with $H = 0$.

Co, Fe, and NiFe and CoFe alloys were tried in this study as both bottom and top electrodes. The best results were obtained with a CoFe bottom electrode and a Co or NiFe top electrode. With Fe films as one of the electrodes, for the thickness range of Al used, the junctions were too low in resistance. Here the results obtained with CoFe/Al$_2$O$_3$/Co or CoFe/Al$_2$O$_3$/NiFe tunnel junctions are presented. The possible presence of a monolayer or so of Al metal left unoxidized, although not indicated in the sample formula, cannot be ruled out. Previous tunneling studies on the magnetic proximity effect in Fe/Au and Fe/Al bilayers show only a small decrease in the spin polarization of the electrons when a normal metal of monolayer thickness is deposited over the Fe [14].

The quality of the tunnel junctions was verified by current-voltage and conductance (G) measurements at 295, 77, and 4.2 K. In Figs. 1(a) and 1(b) the tunnel conductance G is plotted against the applied voltage across the junction for a CoFe/Al$_2$O$_3$/NiFe junction at 295 and 4.2 K. CoFe/Al$_2$O$_3$/Co junctions show identical results. At low bias [Fig. 1(a)] G is nearly independent of the bias up to about ±15 mV, as it should be for a good tunnel junction with an Al$_2$O$_3$ tunnel barrier [1,15]. The dip at $V = 0$ at 4.2 K (but not present at 295 K) is a feature often seen in tunnel junctions whose significance will be discussed below. In the higher bias region [Fig. 1(b)], close to parabolic dependence of G is observed. Such a dependence is regularly seen for high quality Al/Al$_2$O$_3$/FM junctions [1,15]. The overall shape of the conductance is the same at 295 and 77 K also, although the junction resistance increased by tens of percent on going from 295 K to lower temperatures.

The current-voltage data at 295 K were fitted by Simmons' theory of tunneling [16] to obtain values of tunnel barrier height (ϕ) and thickness (d). For the NiFe and Co junctions the values $\phi \approx 1.9$ eV and $d \approx 16$ Å obtained agree well with the standard Al/Al$_2$O$_3$/FM tunnel junction parameters [15]. These data together with the I-V and G-V measurements and their temperature dependence give confidence that tunneling is the major conduction process.

Unlike the conductance measurement, the field dependence of the junction resistance was performed by a four-terminal technique. Figure 2 shows results for a CoFe/Al$_2$O$_3$/Co junction. Replacing Co with NiFe as the top electrode showed similar behavior. The upper two curves show the small magnetoresistance changes in the two films (not the junction) which mark the coercive fields (H_C) by the position of their extrema [17]. (The presence of a maximum or minimum is determined by the field and current directions.) The value of H_C for the films determined from the M-H loop measurement using a SQUID magnetometer agrees with the above conclusion. For junctions, as the magnetic field decreases, R increases slowly. Upon reversing the field, R begins to increase sharply, showing a peak. With further increase in H, the resistance drops quickly and attains a constant value. This behavior is seen for H both parallel and perpendicular to the junction plane. For the latter case, however, the peaks are broader and shifted to higher fields as one would expect in these FM films which have an in-plane easy axis of magnetization. The change in the junction resistance with respect to the absolute value at the peak, $\Delta R/R$, for this junction is 10.6% at 295 K. In several tens of junctions over a 10% effect has been observed at room temperature;

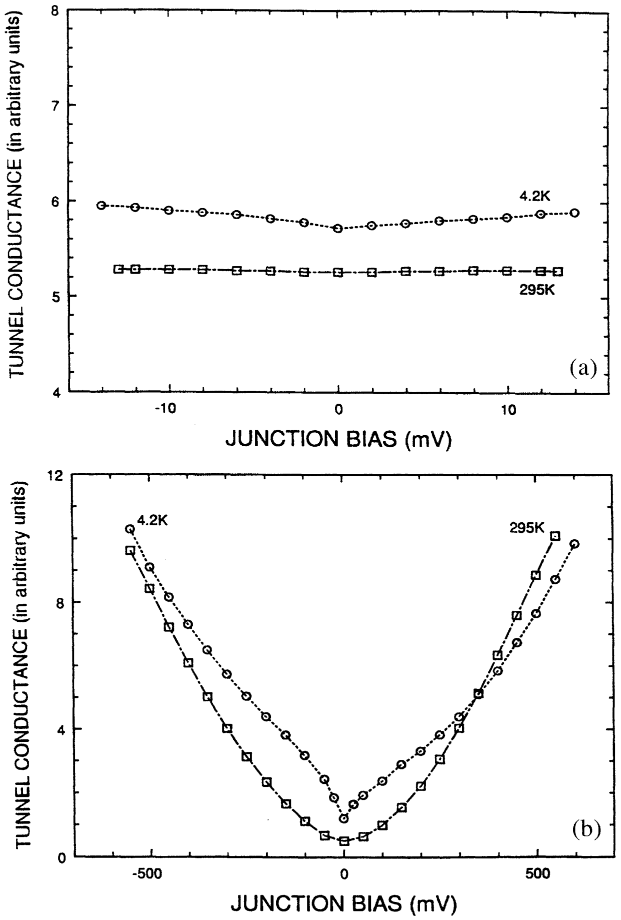

FIG. 1. Tunnel conductance plotted as a function of the applied dc bias for a CoFe/Al$_2$O$_3$/NiFe tunnel junction at 4.2 and 295 K in zero field.

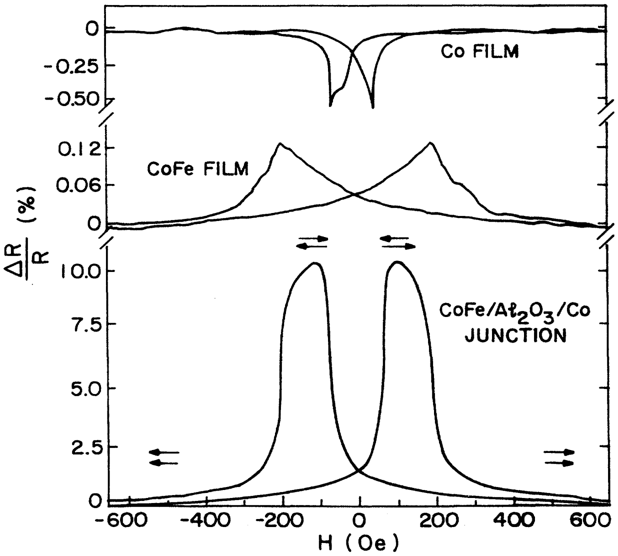

FIG. 2. Resistance of CoFe/Al_2O_3/Co junction plotted as a function of H in the film plane, at 295 K. Also shown is the variation in the CoFe and Co film resistance. The arrows indicate the direction of M in the two films (see text).

some junctions showed values up to 11.8%. This change in R with field is far higher than previous reported values [5–12]. The changes in the resistance for the full length of the strips are 0.6% and 0.1% for Co and CoFe films, respectively, as shown in this figure. Only about $\frac{1}{30}$th of the actual film comprises the junction area, and hence the lead contribution is negligible to $\Delta R/R$ of the junction.

In general, the percentage change of junction resistance nearly doubled at 77 K compared to its value at 295 K. Further increase occurred upon cooling the junction to 4.2 K, reaching values up to about 24% in some cases. As T decreased the resistance peak broadened slightly, and, in addition, there was a small shift in the peak position to higher field values. In a few cases there was little change in H_C for CoFe, whereas the top Co film had a large increase in H_C upon cooling to 77 K, thereby bringing the two coercive forces near to each other. In such samples $\Delta R/R$ at 77 K was nearly the same as or even smaller than the 295 K value.

In order to investigate the effect of dc bias, R vs H for several junctions was studied by using dc current through the junction. Similar results for the H and T dependence of junction resistance were observed. Peak position, peak width, and change with temperature are almost independent of dc bias up to about 100 mV, and there was only a small decrease in $\Delta R/R$ from the value measured by the ac technique. However, at large biases there was a significant decrease in the value of $\Delta R/R$ as shown in Fig. 3. The value of the ratio obtained by the ac technique is plotted on the ordinate. The decrease observed with increasing dc bias is similar at all three temperatures in the low bias region, indicating negligible Joule heating effects.

The field dependence of resistance in FM/I/FM junctions can be explained qualitatively based on earlier models [2,5,13]. At high fields (beyond the H_C of the FM films), the magnetizations of the two FM films are fully saturated and aligned in the field direction. The tunneling probability and hence the current is high. As H decreases toward zero and changes sign, M in the film with lower H_C reverses, whereas for the film which has the higher value of H_C, M remains the same. In this field range, M in the two films are antiparallel to each other, thereby lowering the tunneling probability and tunnel current. Upon raising the field further in the reverse direction, M in the second film also reverses, becoming parallel to the first film and H. This leads again to higher tunneling probability and current. The change in tunnel current (and the junction resistance) depends crucially on the difference of H_C in the FM electrodes. The temperature dependence of H_C can also cause a temperature dependence in the fractional change in resistance. H_C for CoFe, NiFe, and Co films changed by different amounts; Co showing the maximum increase in H_C at lower temperatures. When T decreased, H_C for the two films moved farther apart, thereby allowing the antiparallel alignment to exist in a wider range of H, increasing $\Delta R/R$. One can now understand why the peak in the junction resistance occurs at field values between the peak of the CoFe film and the dip of the Co film (see Fig. 2). When H_C for the two films was nearly the same and the position of the $R(H)$ peaks of the two films nearly overlapped, very small or no change was seen in junction resistance. The temperature dependence of $\Delta R/R$ cannot be caused by a change in M because at thicknesses of about 100 Å these FM films behave nearly like bulk samples, which have Curie temperatures greater than 1200 K, and hence should have negligible effect on M below 295 K.

The fractional change in junction resistance with H can be calculated using Eq. (1), knowing the electron spin polarization of the films. For CoFe, P was measured and

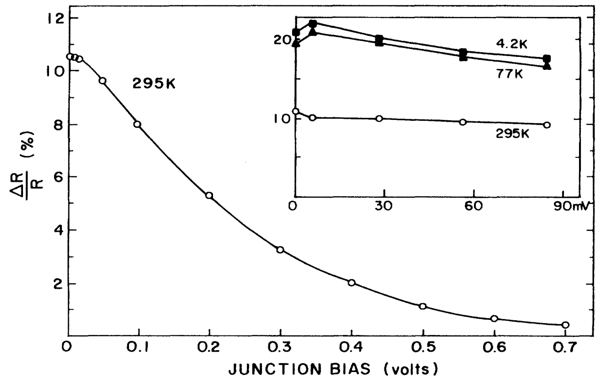

FIG. 3. The ratio of $\Delta R/R$ plotted as a function of dc bias for CoFe/Al_2O_3/Co junction. Inset: Low dc-bias region at three different temperatures. The abscissa in the inset is for 4.2 K data, which are twice the values at 295 K. The increase in $\Delta R/R$ as T decreased is seen in the inset.

found to be 47% [18]. Taking the value of 34% for Co metal [1], Eq. (1) gives 27% change for CoFe/Al_2O_3/Co junctions. The maximum observed value at 4.2 K is 24%, close to the predicted value, although it decreases to 11.8% at 295 K. Before this work, the highest value was only about (2–7)% at 4.2 K and 2.7% at 295 K. The present junction success is partly attributed to a smoother FM film brought about by depositing onto a LN_2-cooled substrate with Si as the nucleating layer underneath the first FM film (known to occur in thin films) and using a thinner FM film. The degree of smoothness of our films is yet to be measured.

The conductance dip at $V = 0$, shown in Fig. 1(b), was observed at 4.2 K and slightly at 77 K, but was absent at 295 K. Various causes of such anomalies have been discussed [15], including localization in amorphous materials or ultrathin films, magnetic scattering in the electrodes or in the barrier, and tunneling through an intermediate state. That the anomaly is not the cause of the large value of $\Delta R/R$ is made clear from the independence of low bias voltage in the region where the anomaly is changing most rapidly.

The decrease of magnetoresistance with dc bias is not understood at present. It is not implied by the simple Julliere model; only a small effect because of the variation of the density of states for a voltage of 0.7 V near the Fermi energy is expected. The Slonczewski theory [13] predicts a decrease in the spin polarization with barrier height, but for the characteristics of the present junctions it seems to predict a value of $\Delta R/R < 1\%$. Another explanation of the decrease in $\Delta R/R$ as the temperature and voltage bias increase is the formation of magnons in the tunnel barrier. Such magnons have been found in NiO by Tsui, Dietz, and Walker [19]. It is reasonable that the glow discharge may oxidize the surface of the first FM electrode to form a thin semiconducting layer. This layer would be consistent with the increased temperature change in junction resistance over that of a normal Al oxide barrier. The small structure on the G-V curves at about 150 mV is consistent with magnon formation and might cause spin flipping in the tunneling process. Detailed T and H dependence would be helpful in elucidating the effect of such a magnetic oxide, if present.

Ferromagnetic-insulator-ferromagnetic tunneling has been shown to give over 10% change in the junction resistance with H less than 100 Oe, at room temperature. The effect is reproducible and the change increases with decreasing temperature but decreases at high dc bias across the junction. Using such junctions as magnetic sensors or memory elements would have several advantages: it is a trilayer device and does not strongly depend on the thickness of FM electrodes or the tunnel barrier; submicron size is possible with high junction resistance and low-power dissipation. The magnitude of the effect is consistent with the simple model of spin-polarized tunneling between ferromagnets. With improved control and characterization of the junction fabrication a quantitative comparison of results with different ferromagnetic metals of known spin polarization would give a rigorous test of proposed theories and determine the origin of the temperature and voltage dependence.

It is a pleasure to thank G. Reynolds for the SQUID measurements and P. LeClair for assisting with the fit to Simmons' theory. L. Kinder thanks NSF for sponsoring the High School Honors Program through Boston University and T. M. Wong thanks MIT for the UROP fellowship. This research was supported by NSF Grant No. DMR-9023400 and ONR Grant No. N00014-92-J-1847.

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