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Resonant tunnelling in Dy- or Gd-doped Al_2O_3 magnetic tunnel junctions

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Abstract

Tunnelling characteristics have been measured for Co–insulator–Py magnetic junctions with Dy- or Gd-doped Al_2O_3 barriers. The theoretically predicted enhancement of the magneto-resistance due to resonant tunnelling in the presence of paramagnetic impurities is not borne out. However, even a full monolayer of Dy or Gd has no detrimental effect on the junction magneto-resistance (JMR) at low temperature and low bias voltage. With increasing temperature and bias, the JMR of the doped junctions decreases significantly faster than the JMR of the Al_2O_3 control junctions. Junctions in which the entire barrier has been replaced by Dy_2O_3 or Gd_2O_3 show strong non-linear current–voltage characteristics, but display no JMR. It is shown that not direct tunnelling but spin-independent impurity-assisted tunnelling is the primary conductance channel in these junctions.

1. Introduction

Magnetic tunnel junctions consist of two ferromagnetic electrodes separated by a thin insulating barrier. The size of the conductance due to direct tunnelling shows a large difference between the parallel G_p and antiparallel G_{ap} alignment of the two magnetic electrodes [1–4]. This effect has been predicted by Julliere [5], who showed that the junction magneto-resistance (JMR) is proportional to the spin polarization P of the two electrodes and is given by $\Delta G/G_p = 2P_1P_2/(1 + P_1P_2)$. It has been remarked recently that even larger values of JMR can be achieved when the tunnelling process is due to resonant tunnelling in the presence of paramagnetic impurities. Based on work of Bratkovsky [6] and Slonczewski [7], Jansen

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and Lodder [8] and Vedyayev *et al* [9] showed that when a sheet of fully aligned paramagnetic impurities is placed within a tunnel barrier, an enhancement of the magneto-resistance (MR) will result. The required alignment of the magnetic impurities is supposedly due to the molecular field of the ferromagnetic electrodes. Vedyayev *et al* [9] placed the sheet of impurities off the centre of the barrier and took a realistic, but arbitrary, value for the molecular field which enabled them to calculate the bias voltage and temperature dependence of the resonant MR. If the sheet of impurities is placed in the centre of the barrier, the impurities will be randomly oriented in the antiparallel alignment due to the competing interaction from both electrodes. It follows however from the calculation of Jansen and Lodder [8] that when the electrodes are antiparallel aligned, the resonant conduction is independent of the alignment of the impurities. The sheet of impurities can therefore be placed in the centre of the barrier without detrimental effects on the theoretical value for the MR.

Experimentally, Jansen and Moodera [10–12] have incorporated impurities in the barrier showing a decrease in MR for Ni, Co and non-magnetic impurities. This has been explained theoretically by Tsymbal and Pettifor [13, 14]. Only for Fe impurities has an increase in MR been detected, a roughly constant enhancement of the MR by a factor of 1.1 to 1.2 at temperatures up to 300 K and bias voltage up to 0.6 V. This lack of variation of the MR enhancement with temperature and bias voltage is in stark contrast to theoretical predictions [9, 15] and indicates that the enhancement may not be due to alignment of the magnetic impurities in the barrier.

In this paper, we shall try to show evidence of resonant tunnel MR by doping Al_2O_3 barriers with rare-earth metals, Dy or Gd. The energy of formation of the rare-earth (R) oxides is larger than for Al oxide, namely >1800 and 1676 kJ mol^{-1} for R_2O_3 and Al_2O_3, respectively [16]. Both Dy and Gd are hence expected to be fully oxidized trivalent ions after the glow discharge. The ions will have a large magnetic moment derived from the open 4f core shell of 10.63 and 7.94 μ_B for Dy and Gd, respectively. With similar bandgap and electron affinity, the barrier height is expected to be of the same order as for Al_2O_3. Hence the rare-earth- doped tunnel junctions provide the ideal system to study resonant tunnelling in the presence of paramagnetic impurities. To further improve understanding of the tunnelling mechanism in magnetic barriers, we also made junctions in which the entire barrier was made up of Dy_2O_3 or Gd_2O_3.

2. Experimental details

Preparation of the tunnel junctions follows the same thermal evaporation process as used by Jansen and Moodera [17] to incorporate dopants in a narrow region of the barrier. Co strips of 8 nm are deposited onto a glass substrate covered with a 1 nm Si seed layer. One half of the Co strips is used to form control junctions of Al_2O_3, the other half to form the R-doped barriers. The R-doped barriers are formed by subsequent evaporation of a 0.6 nm layer of Al, a 0.05–0.25 nm layer of rare-earth element and another 0.6 nm layer of Al. *In situ* glow discharge leads to roughly 1.6 nm Al_2O_3 barriers for the control junctions and 0.8 nm Al_2O_3/up to 0.3 nm R_2O_3/0.8 nm Al_2O_3 barriers for the junction doped with Dy or Gd. The top electrode is formed by cross strips of 10 nm Py ($Ni_{80}Fe_{20}$) and the junction area is approximately 4×10^{-8} m^2.

Junctions with their entire barrier consisting of Dy_2O_3 or Gd_2O_3 have also been made by evaporation of the metallic elements followed by *in situ* glow discharge. MR has been measured using a dc four-terminal method. Cryogenic measurements were performed by immersing the junctions in liquid nitrogen or in liquid helium in a glass Dewar system that could be pumped down to reduce the temperature of the liquid helium to close to 1 K.

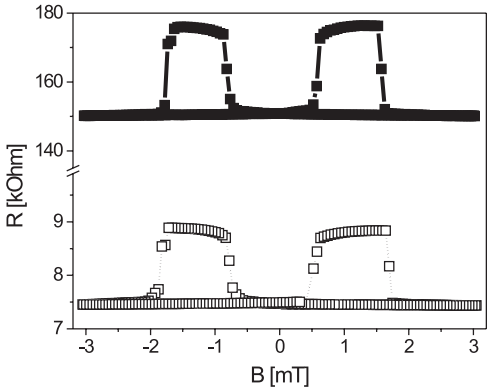

Figure 1. Field dependence of the resistance of a Co/1.2 nm Al_2O_3/Py (open squares) and a Co/Dy-doped Al_2O_3/Py (filled squares) tunnel junction. The amount of Dy in the barrier is 0.25 nm. Measurements are made at 77 K and 10 mV bias voltage.

3. Results

3.1. δ-doped barriers

In figure 1, a typical resistance versus magnetic field curve is shown for a control junction and a junction with a 0.25 nm layer of Dy sandwiched in between two 0.6 nm layers of Al at 77 K and 10 mV bias voltage. The switching fields are determined by the electrodes only and hence are the same in both junctions, 0.8 mT for the Py electrode and 1.7 mT for the Co electrode. The resistance of the Dy-doped junction is a factor of 20 larger than the control junction due to the increased barrier thickness in the Dy-doped junction. Remarkably, the MR is not affected at all by the addition of such a large quantity of Dy. Both the control junction and the Dy-doped junction show 16% JMR at low temperature and low bias voltage. An estimate of the barrier height is obtained by applying Simmons' formula [18] to the conductance at zero bias voltage at low temperature while keeping the barrier thickness fixed at the values that follow from the oxidation as given in the experimental section. This is a better method than fitting the I–V curve since a considerable part of the conductance at higher voltage is not due to direct tunnelling, which leads to unrealistic high values for the barrier thickness as will be discussed below. The average barrier height for the control junctions is 2.0 eV (with barrier width 1.6 nm) and the junctions doped with a maximum amount of Dy have a barrier height of 1.75 eV (at 1.9 nm barrier width). This means that the addition of Dy distorts the barrier height slightly.

In figure 2, the bias dependence of the MR is plotted for the junctions described in figure 1, both at 77 K and at room temperature (RT). The asymmetry in the bias dependence of the MR was of the order of 10% at high bias voltage due to the difference in electrode material. This has been averaged out in the figure. As followed from the previous figure the magnitude of the JMR is equal for the control junction and the Dy-doped junction at low bias and 77 K. However, with increasing bias, the JMR of the Dy-doped junction decreases considerably faster than the JMR of the control junction. Increasing the temperature has a similar effect, with the JMR of the Dy-doped junction considerably lower at RT than the JMR of the control junction, even at low bias. These results are distinct from data previously collected on junctions doped with 3d metals [10, 11, 17]. Fe doping showed an enhancement of JMR independent of temperature, Co showed a decrease of JMR further exaggerated by temperature increase and Ni showed a

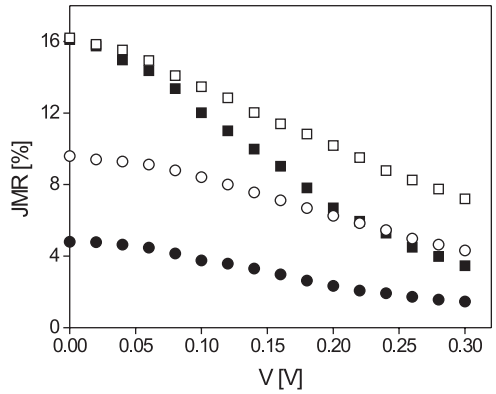

Figure 2. Bias dependence of the MR of the tunnel junctions described in figure 1 (Co/1.2 nm Al_2O_3/Py (open) and Co/Dy-doped Al_2O_3/Py (filled)). Measurements are made at RT (circles) and 77 K (squares). Whereas at low temperature and bias the MR is identical, the doped barrier has a much stronger decrease in MR with increasing temperature and bias.

very strong decrease at all temperatures. These Dy-doped junctions are the only ones to show a different temperature and bias voltage dependence while having the same JMR as the control junctions at low temperature and low bias.

Only results for a junction with 0.25 nm Dy doping have been given above. Junctions with less Dy doping show a similar pattern but with MR nearer to that of the control junction. For reasons of clarity the junctions with maximum Dy doping have been selected for detailed study. The Gd-doped junctions show similar results, but the JMR does not equal the JMR of the control junctions until liquid He temperature has been reached. Reducing the temperature from 4 to 1 K had no further effect on the MR. For both Dy and Gd doping, the RT MR decreases with increasing dopant concentration (although with rather large scatter). For 0.25 nm Gd and Dy doping, the zero-bias RT JMR is approximately 80 and 60% of the JMR value of the control junction, respectively.

3.2. Dy_2O_3 and Gd_2O_3 barriers

Subsequently, tunnel junctions were prepared in which the entire barrier was formed by rare-earth oxides, either 1.5 nm Dy_2O_3 or 1.5 nm Gd_2O_3 (1.2 nm before oxidation). These tunnel junctions did not show MR at any temperature down to 1 K. In figure 3, the static conductance is shown as a function of the squared bias voltage for Al_2O_3, Dy_2O_3 and Gd_2O_3 barriers of the same thickness. This rather unconventional way of plotting is more indicative of the conduction processes than the conventional R–V plot. The parallel conductance plot for the Al_2O_3 junction fits, besides the well known conduction dip near zero bias [1], Simmons' formula for direct tunnelling through a square barrier written in the form $G(V) = \alpha + \gamma V^2$ [18]. Notice that $\gamma V^2 \ll \alpha$ as required from the assumptions leading to Simmons's formula [19–21]. The Dy_2O_3 and Gd_2O_3 barrier junctions fit the same mathematical expression $G(V) = \alpha + \gamma V^2$, but the large dependence on V^2 cannot be due to direct tunnelling. It is therefore not useful to extract a barrier height and thickness from the fit.

The temperature and bias dependence of the Dy_2O_3 and Gd_2O_3 barrier junctions has been measured in detail. At all temperatures, the (spin-independent) conductance could be described accurately by $G(V, T) = \alpha(T) + \gamma(T)V^2$. The variable $\alpha(T)$ increased with temperature but the temperature relation varied strongly for different barriers and different samples with no

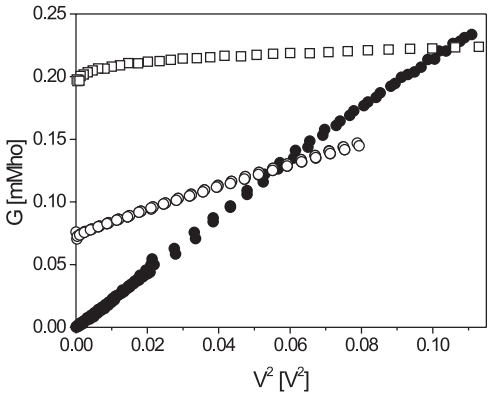

Figure 3. Bias dependence of the conductance at 4.2 K for Co/insulator/Py junctions with 1.5 nm of Dy_2O_3(open circles), Gd_2O_3 (filled circles) and Al_2O_3 (open squares) as barrier. The conductance of the Gd_2O_3 junction at zero bias is smaller than 1 μS.

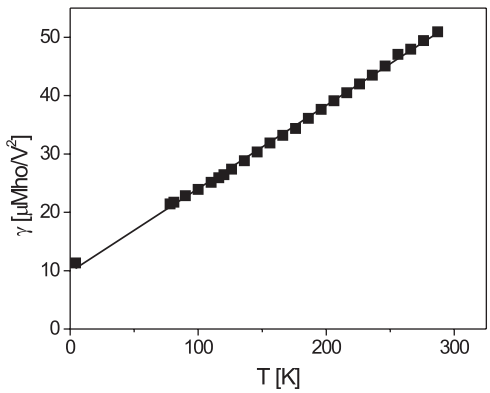

Figure 4. Temperature dependence of the bias dependence parameter γ for a tunnel junction with a 1.7 nm Dy_2O_3 barrier. The line is a linear fit to the data.

clear pattern. On the other hand, the variable $\gamma(T)$ showed a linear dependence on temperature for all junctions. In figure 4, the value of $\gamma(T)$ has been plotted for a junction with a 1.7 nm Dy_2O_3 barrier. The linear fit to $\gamma = \gamma_0 + \gamma_1 T$ gives $\gamma_0 = 10\,\mu\mathrm{S\,V^{-2}}$ and $\gamma_1 = 1.4\,\mu\mathrm{S\,K^{-1}\,V^{-2}}$.

4. Discussion

No JMR has been observed in any junction in which the entire barrier was formed by either Dy_2O_3 or Gd_2O_3. This is in contrast to measurements from Nowak and Rauluszkiewicz [22] and LeClair *et al* [23], which show a few per cent MR at 4.2 K and 100 mT for Fe/Gd_2O_3/Fe and Gd/Gd_2O_3/Py junctions, respectively. Furthermore, 15% spin polarization has been detected for Gd in Gd/Gd_2O_3/Al in even higher magnetic field [24]. It is clear that direct tunnelling is totally suppressed in our Gd_2O_3 junctions ($\ll 1\mu$S at 10 mV, indicating a barrier height of at least 2.5 eV), which explains the absence of MR. However, for the Dy_2O_3 barrier, a considerable residual conductance is present at low temperature and low bias, which is possibly due to direct tunnelling. Absence of JMR might be explained by assuming that the interface between the magnetic Co or Py layer and the rare-earth oxide quenches the polarization of the tunnelling

conduction electrons. The importance of the interfacial density of states has been pointed out recently in many articles [25–27]. The interface with an Fe or Gd layer might give a non-zero polarization, but it is more likely that the high magnetic field in which these (pre-1995) junctions were measured is responsible for the discrepancy.

The G–V curves of the Dy_2O_3 or Gd_2O_3 barrier junctions show the existence of a impurity-assisted channel for tunnelling which depends quadratically on the bias voltage and increases with increasing temperature. The same mechanism can be invoked to explain the data on the Dy- or Gd-doped Al_2O_3 barrier junctions. At low temperature and low bias voltage the impurity-assisted tunnelling current is small compared with the direct tunnelling current and the JMR of the Dy- or Gd-doped Al_2O_3 junctions is equal to the JMR of the control Al_2O_3 barrier junctions. Increasing temperature and bias voltage will increase the relative importance of this channel compared with direct tunnelling in the Dy- or Gd-doped Al_2O_3 junctions and hence result in a stronger decrease of JMR with temperature and bias voltage in these junctions than in the control junctions. The exact origin of this conduction channel is not clear at the moment, but its reproducible temperature and bias voltage dependence fitting the expression $G(V, T) = \alpha(T) + (\gamma_0 + \gamma_1 T)V^2$ should provide some clues for future theoretical work.

In the Dy- or Gd-doped barriers the impurity-assisted tunnelling channel does not lead to enhanced MR as predicted in the theoretical articles cited in the introduction. The most obvious explanation for this is the absence of alignment of the magnetic moments. In [9] it was assumed that the alignment of the magnetic moments is due to the superexchange between the bulk ferromagnetic layer and the magnetic impurities. In our experiments the expected exchange field is considerably lower because the distance between the layers is 0.8 nm compared with 0.4 nm in the theoretical description (experimentally such a thin layer would lead to pinholes and uncertainty about the interface composition). However, even when spin-disorder is frozen out at 1 K, no enhancement of JMR is detected. It might be that a too optimistic estimate of the value of the superexchange has been made or that the rare-earth–rare-earth interaction within the layer could not be ignored. (Bulk Gd_2O_3 and Dy_2O_3 order anti-ferromagnetically at a few kelvin, but the tunnel barriers are most probably amorphous.) Another, almost trivial, explanation for the absence of enhancement would be that the value of the MR for resonant tunnelling is very similar to the MR value due to direct tunnelling.

In conclusion, the predicted enhancement of the MR for barriers doped with paramagnetic impurities has not been shown experimentally. The absence of alignment of the dopants seems to be the underlying cause. The dopants have, however, no detrimental effect on the MR at low temperature and low bias voltage. Data on the doped Al_2O_3 barriers and the entire Gd_2O_3 or Dy_2O_3 barriers indicate the existence of a single spin-independent conduction channel besides direct tunnelling. The increase in the relative importance of this conduction channel is responsible for reducing JMR in the doped junctions at high temperatures and high bias voltage. The absence of polarization at the metal/rare-earth interface is most likely responsible for the lack of MR in the rare-earth oxide barriers.

Acknowledgments

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