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Spin-polarized tunnelling, magnetoresistance and interfacial effects in ferromagnetic junctions

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ABSTRACT

The pioneering studies of spin-polarized tunnelling by Meservey and Tedrow in the early 1970s showed that the conduction electrons in ferromagnetic (FM) metals are spin polarized and that the spin is conserved in the tunnelling process. Only recently (1995) improved material fabrication techniques have permitted realization of the Jullière quantitative model, showing that tunnelling in ferromagnet/insulator/ferromagnet (FM/I/FM) junctions should lead to a large junction magnetoresistance (JMR); JMR values greater than 30% have been achieved at room temperature. This recent success has led to several fundamental questions regarding the phenomenon of spin tunnelling and also the development of JMR devices. In this paper, experimental results, such as the dependence on bias, temperature and barrier characteristics of FM/I/FM tunnelling are reviewed briefly. The influence of inelastic tunnelling processes, metal at the interface and material properties on the JMR is discussed. The future direction from both the physics and the applications viewpoints, is also covered.

§1. INTRODUCTION

Spin-polarized tunnelling (SPT), discovered by Meservey *et al.* (1970) and Merservey and Tedrow (1971, 1994), laid the foundation to a new field of research. Meservey and Tedrow measured the conduction-electron spin polarization P in magnetic metals and compounds using the Zeeman split quasiparticle density of states in a superconductor as the spin detector. Tunnelling from a ferromagnetic (FM) film, with its uneven spin distribution at the Fermi level E_F, into such a spin-split superconducting Al film reflects the spin polarization of the tunnelling electrons coming from the ferromagnet. Values, of P recently measured are higher owing to improved junction preparation conditions including samples grown by molecular-beam epitaxy. Highly polarized tunnelling electrons can also be obtained through a phenomenon called the spin-filter effect using magnetic semiconductors such as EuS and EuSe as tunnel barriers (Moodera *et al.* 1988, 1990, 1993).

Jullière (1975) made the first reported magnetoresistance measurement on a ferromagnet/insulator/ferromagnet (FM/I/FM) trilayer junction and interpreted it by stating that the tunnelling current should depend on the relative orientation of the magnetizations of the electrodes. The tunnel junction magnetoresistance (JMR) is

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defined in this model as

$$\text{JMR} = \frac{\Delta R}{R} = \frac{R_A - R_P}{R_A} = \frac{2P_1P_2}{1 + P_1P_2}, \quad (1)$$

where P_1 and P_2 are the spin polarization of the two FM electrodes, and R_A and R_P represent the junction resistances when the two FM layers have their magnetizations M antiparallel and parallel respectively. This elegant model by Jullière, as we shall see later, turns out to be quite good in predicting the magnitude of JMR seen in clean junctions.

Although tunnelling between two FM films appears to be simple, yet it was not successfully realized for 20 years. Several factors contributed to the failed attempts by many groups, and success still continues to elude many. The major problems are related to the surface roughness of the FM electrodes, the tunnel barrier, the interface quality, the nature of the FM electrodes and the domain walls.

The success in observing a large magnetoresistance in FM/I/FM tunnel junctions happened in 1995 by carefully addressing most of the problems mentioned above and thus obtaining a greater than 10% JMR consistently (Moodera *et al.* 1995). The purpose of this article is to highlight the phenomenal development in this area in the last 4 years.

§ 2. Ferromagnet/insulator/ferromagnet tunnel junctions

Magnetic tunnel junctions (MTJs) were prepared *in situ* by thermal evaporation. Cryogenic evaporation through shadow masks is utilized to create a cross-geometry junction structure of area $(4\text{-}6) \times 10^{-4}$ cm^2. In general, the first FM film (a long strip 80 Å thick and 0.2 mm wide on a Si seed layer) is deposited on a liquid-N_2-cooled glass substrate. To create the tunnel barrier, 8–16 Å of Al film is deposited over it, which is subsequently oxidized at room temperature using O_2 plasma. Cross-strips of the top FM film, 100–200 Å thick and 0.2–0.3 mm wide were then deposited as the second electrode. FM films were grown in an applied field of about 100 Oe.

R_J as a function of H for a $Co/Al_2O_3/Ni_{80}F_{20}$ junction with 12 Å of Al_2O_3 is shown in figure 1 (Moodera *et al.* 1998). The JMRs seen in this case (defined with respect to the peak resistance) are 20.2% and 27.1% at 295 and 77 K respectively,

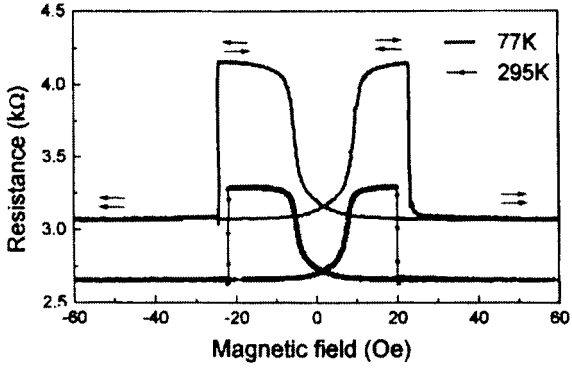

Figure 1. Resistance versus applied magnetic field for a $Co/Al_2O_3/Ni_{80}Fe_{20}$ junction at room temperature and 77 K, showing JMR values of 20.2% and 27.1% respectively. The barrier is formed by oxidation of a 8 Å Al layer. (After Moodera *et al.* (1998).)

changing to 27.3% upon cooling to 4.2 K. With a Co–Fe electrode, at 77 K, a JMR of 32% has been observed (Moodera *et al.* 1997b). The peak value of R_J can be maintained with H turned to zero, leading to two stable states of resistance at $H = 0$, even in the absence of a dc bias, thus giving a non-volatile two-level memory state.

The R_J versus H curve can be understood on the basis of the Jullière model (direction of M indicated by the arrows in figure 1). Taking $P_{Co} = 35\%$ and $P_{Ni_{80}Fe_{20}} = 45\%$, the Jullière model gives a JMR of 27.2%, in very good agreement with the measured values at low temperatures. Note that the JMR values apparently in excess of the Jullière's model predictions that have been reported (Lu *et al.* 1998) could arise because they referred to earlier unoptimized values of P (Meservey and Tedrow 1994). The Jullière model still sets an upper limit for the JMR when adopting the updated polarization values.

JMR effects have been investigated with a number of FM electrodes including Co, Co–Cr, Co–Fe, $Fe_{0.7}Pt_{0.3}$ and $Ni_{80}Fe_{20}$, and tunnel barriers of Al_2O_3, AlN and MgO. With Co–Fe layers at both the interfaces, Sun *et al.* (1998) have recently reported 27%, one of the highest JMRs at room temperature. Based on the polarization of $P_{Co\text{-}Fe} = 55\%$, the highest P value among the transition-metal ferromagnets, one can expect a JMR of 46% for a good (Co-Fe)/Al_2O_3/(Co-Fe) junction.

The possibility of seeing a high JMR, its stability, the bias dependence and the junction resistance critically depend on the quality of the tunnel barrier. Recent realization occurred with Al_2O_3 barriers (which had been successfully used in SPT experiments in the past (Meservey and Tedrow 1994)) formed by oxidizing a thin Al layer. This technique can also be used for other barrier materials such as Mg and Ta (Gallagher *et al.* 1997, Moodera *et al.* 1997b, Plaskett *et al.* 1997). For FM/I/FM tunnelling the most successful barrier materials until now have been Al_2O_3, AlN and MgO, whereas other barriers that have been tried are in general non-stoichiometric and/or magnetic (Platt *et al.* 1996, 1997). These latter barriers can lead to spin memory loss or spin scattering (see section 5).

In general, for uniform coverage the Al film thickness ranged from about 7 to 18 Å, depending also on the type of FM electrode (Moodera *et al.* 1997a, R. van de Veerdonk and J. S. Moodera, unpublished). There is a small range of Al thicknesses that yield the best JMR for a given oxidation condition. With thinner Al, the uncovered FM surface will become oxidized during barrier formation. On the other hand, with too thick an Al film, excess Al metal will be left behind unoxidized, reducing the polarization (Moodera *et al.* 1989) and hence the JMR. One interesting observation was that, even with 4 Å Al coverage, a JMR of 10% was seen. Uniformity and its stoichiometry of Al_2O_3 have been characterized using Rutherford back scattering (RBS) and X-ray photoelectron spectroscopy (Bobo *et al.* 1998, Sun *et al.* 1998, Matsuda *et al.* 1999).

When the actual junction resistance R_T becomes comparable with the resistance R_L of the lead over the junction area, current flow becomes non-uniform over the junction area, giving rise to spurious measured junction resistances (Petersen and Vernon 1967, van de Veerdonk *et al.* 1997). In extreme cases, a negative four-terminal dc resistance can be observed. This was qualitatively attributed to the measuring cross-geometry artefact. This geometrical effect, which showed erroneously rather a large value of JMR in macroscopic millimetre sized junctions (Miyazaki and Tezuka 1995), was absent in a later report by Kamugai *et al.* (1997).

§ 3. Bias voltage dependence of the junction magnetoresistance

Tunnel junctions are nonlinear elements; at low biases (much less than the barrier height), they are Ohmic whereas, at higher biases they have nonlinear current–voltage (I–V) characteristics (Wolf 1985). The dynamic conductance G versus the dc bias has nearly a parabolic dependence. However, with a FM electrode, G versus V_{dc} curves can deviate noticeably. For a $Co/Al_2O_3/Ni_{80}Fe_{20}$ junction, the dynamic conductance variation with V_{dc} is asymmetric, which is a common feature for dissimilar metal electrodes. The presence of metal particles, magnons, magnetic impurities, localization effects, multistep tunnelling and states in the barrier or at the interface can adversely affect the spin polarizations of the tunnelling electrons by causing spin-flip scattering (Appelbaum 1967).

Lu *et al.* (1998) saw in their study of dynamic resistance a cusp-like feature at zero bias at lower temperatures, limited to less than 100 mV. They suggested that caused most of the increase in the JMR at zero bias, whereas the decrease with increasing V_{dc} was due to magnon excitations at the FM–I interface. Inelastic tunnelling (IET) spectra measured in zero H at various temperatures (figure 2) showed a peak (dip) at about ± 100 mV and an additional sharp feature at about 17 meV at lower temperatures (which was present even in junctions where only one electrode was magnetic) (Moodera *et al.* 1998). For magnetic junctions, these peaks in the IET spectra have been attributed to magnons generated in the magnetic barrier (Tsui *et al.* 1971) or in FM electrodes during the tunnelling process (Moodera *et al.* 1995, 1998).

Irrespective of the junction quality, the JMR shows a significant decrease with increasing V_{dc} at all temperatures (Moodera *et al.* 1995, 1998, Beech *et al.* 1996, Lu

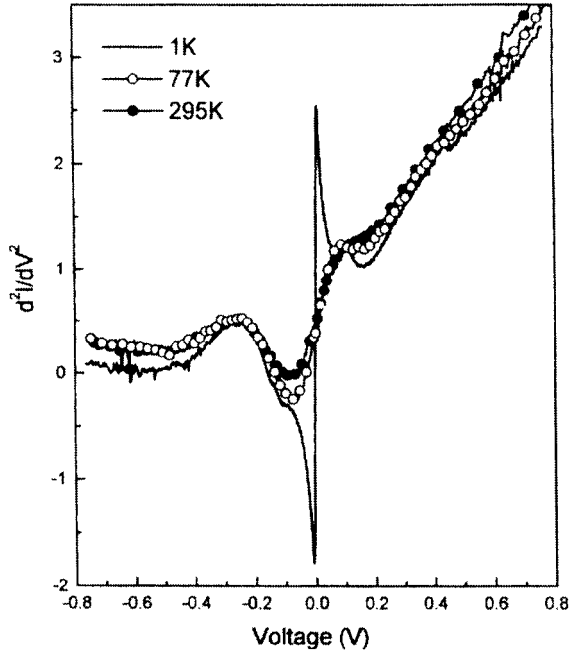

Figure 2. IET spectra at three temperatures for the same junction as in figure 3, measured at $H = 0$. Similar spectra are seen for junctions where one electrode is a ferromagnet and the other electrode is Al. (After Moodera *et al.* (1998).)

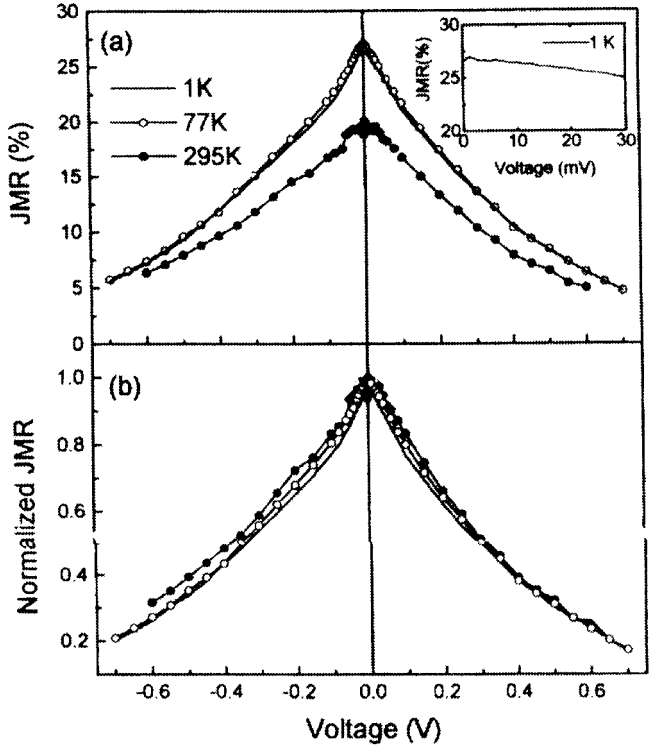

Figure 3. JMR versus dc bias at three temperatures for the same junction as in figure 3. Data shown are (*a*) the actual percentages and (*b*) normalized value at zero bias. The inset shows the JMR in the low-bias region, displaying the near constancy of JMR. (After Moodera *et al.* (1998).)

et al. 1998, Sun *et al.* 1998, Tezuka and Miyazaki 1998). The bias dependence of the JMR at 295, 77 and 1 K for the junction in figure 3 shows a monotonic decrease in the JMR as V_{dc} increases. The normalized data (figure 3 (*b*)) show a temperature independence. The magnitude of the decrease depended not only on the quality of the interfaces and barrier type but also on the FM electrode. Doped junctions (see section 5) or junctions where two-state tunnelling is favoured by the presence of defect states in the barrier (Zhang and White 1998) also showed an increased JMR dependence on V_{dc}. In contrast, in the undoped best junctions the JMR decreases only to about half the value at 0.5 V. Moreover it was observed that the junctions with Ni or $Ni_{80}Fe_{20}$ electrodes showed a stronger decrease in the JMR than junctions with Co or Co–Fe electrodes.

The dc bias dependence of the JMR is not well understood. This has been attributed to several factors: the increase in the conductance with bias, excitation of magnons and the energy dependence of spin polarization due to band-structure effects (Moodera and Kinder 1996, Lu *et al.* 1998, Moodera *et al.* 1998). Recent calculations show that a significant part of the JMR decrease can be attributed to magnon excitation (Bratkovsky 1997, Zhang *et al.* 1997), as also is seen from the IET spectra (Moodera *et al.* 1998). In a later paper, Bratkovsky (1998) provided a model that better fitted the JMR versus V_{dc} data of J. Nickel, T. Anthony and J. A. Brug (1998, unpublished) by including phonon contributions in addition to elastic tunnelling and magnon processes.

§ 4. Annealing effects and temperature dependence

The effect of annealing on R_J, the JMR and the barrier parameters has been investigated by several groups (Sato and Kobayashi 1997, Sun *et al.* 1998). In general the optimum annealing temperature was found to be around 230°C (beyond which the junctions began to deteriorate) to achieve the maximum JMR. The improvement in the junction properties upon annealing has been attributed to barrier homogenization as seen in the RBS analysis of the O distribution (Sousa *et al.* 1999) and better magnetic properties of the FM film near the interface. At much higher temperatures, diffusion of the metal atoms into the barrier can occur, leading to degradation of the junction properties.

The observed JMR in MTJs has, at low temperatures, reached nearly the optimum values expected from the Jullière model, whereas it is lower at higher temperatures. The T dependence of R_J is found not only for MTJs but also for standard junctions with non-magnetic (NM) electrodes. This fact suggests a NM origin of the R_J versus T behaviour. To explain this, the Jullière model has been modified by assuming that, in addition to the conductance due to direct elastic tunnelling, a second conductance G_{SI} to be present, which is taken as unpolarized and hence independent of the relative orientation of M (Shang *et al.* 1998). Then the total conductance is

$$G(\theta) = G_T(1 + P_1P_2\cos\theta) + G_{SI}, \tag{2}$$

where θ is the angle between the directions of M in the two electrodes, for example, $\theta = 0^\circ$ for parallel magnetizations and $\theta = 180^\circ$ for antiparallel magnetizations, and G_T is the pre-factor for direct elastic tunnelling with a temperature dependence of a few per cent between 4.2 K and room temperature as per the theory (Stratton 1962), arising from the broadening of the Fermi distributions in the electrodes. G_{SI} was assumed to be T dependent in a manner determined by the physical mechanism responsible.

Conventionally, values for P are determined from a tunnelling measurement at low temperatures ($T < 1$ K). For alloys, the observation that P scaled approximately with the magnetic moment of the alloy led to the assumption that P varied with T as does the magnetization (Meservey *et al.* 1976, Mathon and Ahmed 1988, MacDonald *et al.* 1998). The change ΔG in the conductance for parallel and antiparallel orientations plotted against T (where ΔG is proportional to P_1P_2 and assuming that $P(T) = P_0(1 - \alpha T^{3/2})$), shown in figure 4, directly reflected the T dependence of P_1 and P_2, indicating a substantial reduction in P. For the Co–Co junction, ΔG showed a much weaker decay compared with junctions having $Ni_{80}Fe_{20}$ as one electrode. Thus, the spin-wave-related reduction in P was larger for $Ni_{80}Fe_{20}$, showing consistency with the T_C for Co and $Ni_{80}Fe_{20}$.

The material-dependent constant α is generally larger for the surface owing to surface exchange softening (Mathon and Ahmed 1988). It has also been observed that both P_0 and α are very sensitive to surface contamination (Pierce *et al.* 1982, Mauri *et al.* 1988). Higher contamination at the interface can lead to higher α, resulting in a considerable decrease in P with increasing T. Valuable insight into these phenomena is expected to be obtained from T-dependent measurements in MTJs, complementing other methods for determining surface magnetic properties, while at the same time providing input for theoretical work aimed at relating P to the intrinsic properties of the FM materials.

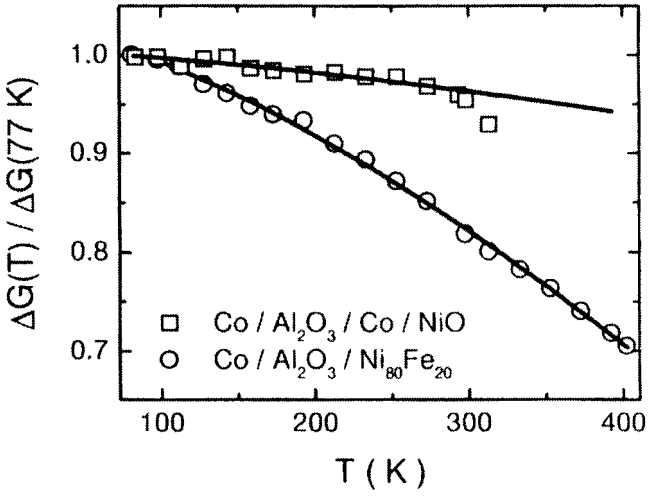

Figure 4. Temperature dependence of the normalized ΔG for two FM junctions: (——) fits to the theory based on thermal spin-wave excitations (Shang *et al.* 1998).

The spin-independent conductance G_{SI} as a function of T showed a $G_{SI}(T) \propto T^{\gamma}$ power-law dependence with $\gamma = 1.35$. Spin-independent contributions can come from imperfections in the Al_2O_3 barrier to provide a noticeable hopping conductance through the associated localized states because of the amorphous character of the Al_2O_3 insulator. Theoretical work (Glatzmann and Matveev 1988) has shown that hopping through chains of N localized states should have a power-law dependence on T, the exponent being $\gamma(N) = N - 2/(N+1)$. The temperature dependence originates from phonon emission or absorption at the transition from the first to the next localized chain. For $N = 2$, $\gamma = 1.33$, close to the experimental value.

§ 5. Barrier doping effects

Magnetic junctions allow us to study electron spin scattering in a systematic and controlled manner, that is by introducing a well defined amount of known foreign elements into the barrier. Note that scattering at E_F is of importance here, as tunnelling electrons generally originate from states in a narrow energy interval around the Fermi level. For that purpose, $Co/Al_2O_3/Ni_{80}Fe_{20}$ junctions prepared with submonolayer amounts of dopants incorporated into the middle of the insulating oxide were studied (Jansen and Moodera 1998).

When a spin-flip event occurs in the barrier, a spin-up electron tunnelling from the FM1 layer has to enter a spin-down empty state in the FM2 layer. In other words, for electrons that change their spin during tunnelling, it is as if the magnetization of electrode 2 has been reversed, that is they exhibit an inverse JMR. Denoting their fraction by f, the conductance for parallel M becomes $(1-f)G_P + fG_{AP}$ and similarly for the antiparallel case, leading to

$$\mathrm{JMR} = \frac{(1-2f)\,\mathrm{JMR}^0}{1 - f\,\mathrm{JMR}^0}, \tag{3}$$

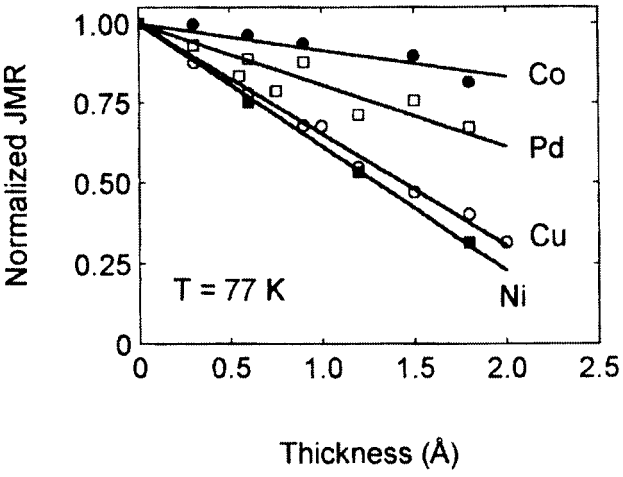

Figure 5. Normalized JMR versus thickness t of the layer of impurities present in the tunnel barrier. Data, measured at 77 K, are shown for Co (●), Pd (□), Cu (○), and Ni (■), together with a linear fit (——) (Jansen and Moodera 1998).

where JMR^0 is the junction magnetoresistance in the absence of spin-flip scattering ($f = 0$). In a first approximation, the JMR is thus expected to decrease linearly with the fraction f.

Dopants such as Ni, Co, Pd, Au and Cu investigated at the submonolayer level showed a significant reduction in the JMR with increasing dopant content. Figure 5 shows the nearly linear decrease in JMR with increase in the dopant thickness for various dopants, Co showing the weakest suppression compared with even Cu or Au (not shown). The linear dependence as expected by equation (3) assumed that the dopant-covered junction area increased linearly with increasing thickness t and also that the fraction f of tunnelling electrons that experience spin flip scales linearly with t. The weak influence of Co was ascribed to the dominant presence of Co^{3+} with no magnetic moment, whereas Ni and Cu ions were in an oxidation state with a magnetic moment. These results agreed with other studies of these ions in Al_2O_3 matrix. Thus incorporation of a submonolayer level of dopants in the barrier in MTJs leads to severe reduction in the JMR as a result of spin scattering. In other words, anything short of single-step tunnelling appears to reduce the JMR.

§6. Magnetic tunnel junctions with non-magnetic interface layers and inverse junction magnetoresistance effects

The basic phenomenon of spin transport through a normal metal layer is not well explored. One can utilize MTJs for such studies. Inserting an ultrathin layer of a NM metal layer at the FM–I interface in a MTJ drastically decreases the JMR, irrespective of the metal (NM layer of Ag, Al, Au, Cu, Pd or Pt) used as shown for some elements in figure 6. In all cases, the JMR reached negligible values with just a few monolayers of the NM layer at the interface. Similar effects were observed by Sun and Frietas (1999) with Cu at the interface. These results are consistent with our earlier direct SPT measurement of the polarization through Au layers using a superconducting Al film (Moodera *et al.* 1989). In other words, these observations show the surface sensitivity of tunnelling as well as the rapid decrease, due to dilution

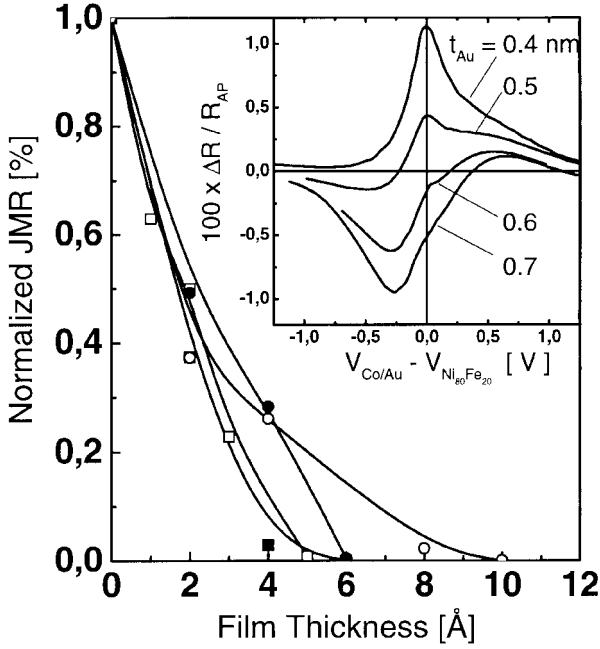

Figure 6. Normalized JMR versus thickness t of the layer of different normal metal impurities at the interface of $Co/M/Al_2O_3/Ni_{80}Fe_{20}$ junctions. Data, measured at room temperature, are shown for Ag (■), Au (□), Pt (●) and Cu (○) with lines to guide the eye (——). The inset shows spin transport through a normal metal: dependence of the JMR on the bias voltage for increasing thickness of the Au interface layer in a $Co/Au/Al_2O_3/NiFe$ tunnel junction at 77 K for $t_{NM} \geqslant 0.4$ nm (Moodera *et al.* 1999).

effects, in the polarization which is induced in the NM layer by the underlying FM layer.

However, in $Co/Au/Al_2O_3/Ni_{80}Fe_{20}$ junctions, for a Au (in some cases also for Cu) film thickness in the range 5–8 Å, a negative JMR effect and an unexpected bias voltage dependence was observed, as shown in figure 6 (Moodera *et al.* 1999), Theoretical calculations by Vedyayev *et al.* (1997) and Zhang and Levy (1998) had predicted oscillations of JMR in FM/NM/I/NM/FM systems as a function of the normal (NM) metal thickness, the interface layer behaving like a quantum well leading to the formation of quantum well states (QWSs) when a resonance condition was fulfilled. Also, according to calculations by Zhang and Levy, the JMR suppression length in the NM layer could be as much as even 100 Å when it was flat whereas, for a rougher FM–NM interface, the coherence was broken, thereby reducing the JMR more rapidly with increasing NM layer thickness. However, in interpreting the experimental results, one has to pay attention to the possibility of interfacial mixing of the atoms (especially in sputtered samples, e.g. Co/Cu), which would yield a spurious decay length. Numerical calculations by our group for the presence of QWSs, based on a model first proposed by Slonczewski (1989), qualitatively explained the experimental features, including its bias dependence (Moodera *et al.* 1999). Such studies may allow one to engineer a special electrode with strong spin filtering, for example by choosing a FM/NM/FM trilayer electrode with a suitable NM layer thickness.

Inverse JMR effects were also observed by Sharma *et al.* (1999) in $Ni_{80}Fe_{20}$ electrodes tunnel junctions with Ta_2O_5 and by de Teresa *et al.* (1999) in $Co/SrTiO_3/La_{0.7}Sr_{0.3}MnO_3$ junctions. They reported a strong bias dependence of the JMR, which was also polarity dependent. The JMR even changed sign with the dc bias, whereas with Al_2O_3 barriers the JMR was positive at all bias voltages. These features have been attributed to the band-structure effects in Ta_2O_5 and $Ni_{80}Fe_{20}$ resulting in the negative polarization at the Ta_2O_5–electrode interface. The main assumption that Sharma *et al.* made to explain their data is different band features and E_F for $Ni_{80}Fe_{20}$ at the interface of Al_2O_3 or Ta_2O_5. They also pointed out the dependence of the inverse JMR magnitude on the barrier oxidation time, which seems to indicate some role played by the barrier defects in these observations, for instance spin-flip scattering.

§ 7. Application potential

Several main areas of possible application for JMR devices are non-volatile magnetic random access memory elements, read head sensors, large arrays of sensors for imaging and ultraflow-field sensors (Prinz and Hathaway 1995, Daughton 1997, Prinz, 1998). Some of the advantages of JMR elements over others are the large signal as well as sensitivity, a non-volatile memory, better radiation hardness and inherent small size. Other issues are dielectric breakdown, noise, long-term stability and switching times. Considerable work is going on in this area (Gider *et al.* 1998, Koch *et al.* 1998, Nowak *et al.* 1998). SPT on an atomic scale is still in an early stage to be technologically viable or even useful for fundamental studies. Among the various approaches, using optically pumped GaAs tip enables spin-polarized vacuum tunnelling and thus imaging of magnetic domain structure of FM films (Alvorado and Renaud 1992, Prins *et al.* 1996). Another approach was chosen in the use of the exchange-split surface state of a ferromagnet to study surface magnetism by SPT (Wiesendanger *et al.* 1990, Bode *et al.* 1998). There is great promise in these techniques.

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