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Spin-Dependent Tunneling in Magnetic Tunnel Junctions

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PACS: 72.25.-b Spin polarized transport
73.40.Gk Tunneling
73.40.Rw Metal-insulator-metal structures
75.70.-i Magnetic properties of thin films, surfaces, and interfaces
85.75.Ss Magnetic field sensors using spin polarized transport
85.75.Dd Magnetic memory using magnetic tunnel junctions

Abstract

The phenomenon of electron tunneling has been known since the advent of quantum mechanics, but continues to enrich our understanding of many fields of physics, as well as creating sub-fields on its own. Spin-dependent tunneling in magnetic tunnel junctions (MTJs) has recently aroused enormous interest and has developed in a vigorous field of research. The large tunneling magnetoresistance (TMR) observed in MTJs garnered much attention due to possible applications in non-volatile random access memories and next-generation magnetic field sensors. This led to a number of fundamental questions regarding the phenomenon of spin-dependent tunneling. In this review article we present an overview of this field of research. We discuss various factors that control the spin polarization and magnetoresistance in magnetic tunnel junctions. Starting from early experiments on spin-dependent tunneling and their interpretation, we consider thereafter recent experiments and models, which highlight the role of the electronic structure of the ferromagnets, the insulating layer and the ferromagnet/insulator interfaces. We also discuss the role of disorder in the barrier and in the ferromagnetic electrodes and their influence on TMR.

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Contents

1. Introduction

In the past few years magnetic tunnel junctions (MTJs) have aroused considerable interest due to their potential applications in spin-electronic devices such as magnetic sensors and magnetic random access memories (MRAMs). The diversity of physical phenomena, which govern functioning of these magnetoresistive devices, makes MTJs also very attractive from the fundamental physics point of view. This stimulated tremendous activity in the experimental and theoretical investigations of the electronic, magnetic and transport properties of MTJs.

A magnetic tunnel junction consists of two ferromagnetic metal layers separated by a thin insulating barrier layer. The insulating layer is so thin (a few nm or less) that electrons can tunnel through the barrier if a bias voltage is applied between the two metal electrodes across the insulator. The most important property of a MTJ is that the tunneling current depends on the relative orientation of the magnetizations of the two ferromagnetic layers, which can be changed by an applied magnetic field. This phenomenon is called tunneling magnetoresistance (sometimes referred to as junction magnetoresistance). Although the tunneling magnetoresistance (TMR) has been known from the experiments of Julliere [1] for almost 30 years, only a relatively modest number of studies have been performed in this field until the mid-nineties. Partly this was caused by technologically demanding fabrication process, which makes it difficult to fabricate robust and reliable tunnel junctions. Also the fact that the reported values of TMR were small (at most a few percent at low temperatures) did not trigger considerable interest in view of sensor/memory applications. A few years ago, however, Miyazaki and Tezuka demonstrated the possibility of large values of TMR in MTJs with Al_2O_3 insulating layers [2], and Moodera *et al*. developed a fabrication process, which appeared to fulfill the requirements for smooth and pinhole-free Al_2O_3 deposition [3]. Since the first observation of reproducible, large magnetoresistance at room temperature, shown in Fig.1, there has been enormous increase of research in this field. Nowadays MTJs that are based on 3d-metal ferromagnets and Al_2O_3 barriers can be routinely fabricated with reproducible characteristics and with TMR values up to 50% at room temperature, making them suitable for industrial applications (see, e.g., ref.[4]).

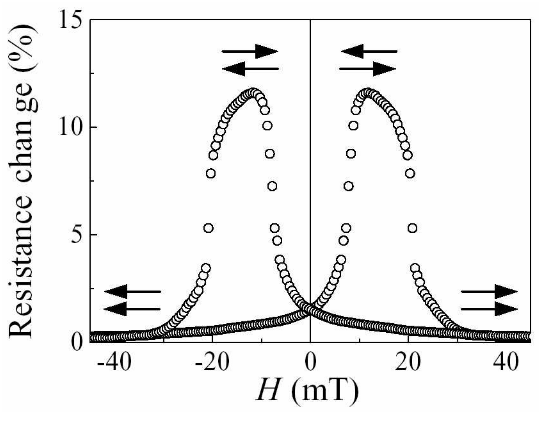

Fig.1 First observation of reproducible, large room temperature magnetoresistance in a $CoFe/Al_2O_3/Co$ magnetic tunnel junction. The arrows indicate the relative magnetization orientation in the CoFe and Co layers. After Moodera *et al*. [3].

TMR is a consequence of spin-dependent tunneling (SDT). The essence of SDT is an imbalance in the electric current carried by up- and down-spin electrons tunneling from a ferromagnet through a tunneling barrier. The origin of this phenomenon can be explained by the fact that the probability for an electron to tunnel through the barrier depends on its Fermi wavevector. In ferromagnetic metals electronic bands are exchange-split, which implies a different Fermi wave vector for the up- and down-spin electrons and consequently a tunneling probability that depends on spin. The spin-dependent tunneling effect was discovered in pioneering experiments by Tedrow and Meservey [5]. Using superconducting layers as detectors they measured the spin polarization of the tunneling current originating from various magnetic electrodes across an alumina barrier. An excellent review on SDT is published by Meservey and Tedrow, which covers the field up to 1994 [6].

The relationship between SDT and TMR was explained by Julliere within a simple model [1] that quantifies the magnitude of TMR in terms of the spin polarizations (SP) of the ferromagnetic electrodes as measured in the experiments on superconductors [5]. Although Julliere's model served as a useful basis for interpreting a number of experiments on TMR, this model is too simple to describe all the available experimental data. In particular, Julliere's model assumes that the SP of the tunneling current is determined solely by the SP of the total electronic density of states (DOS) of the ferromagnetic layers at the Fermi energy. Although later Stearns improved this understanding by considering only the DOS of itinerant electrons [7], the interpretation of TMR in terms of the intrinsic properties of the ferromagnets constituting the MTJ remained unchanged. Experimental results show, however, that the tunneling SP strongly depends on the structural quality of MTJs. Improvements in the quality of the alumina barrier and the metal/alumina interfaces resulted in the enhancement of the measured values of the SP. For example, the SP of permalloy of 32% was obtained in first experiments on tunneling to superconductors [6], but later Moodera *et al*. using improved deposition techniques reported the value of 48% (see ref.[8]). Experiments also show that the SP depends on the choice of the tunneling barrier. Fert and his group found that Co exhibits a negative value of the SP when tunneling occurs through a $SrTiO_3$ barrier [9]. This is opposite to the spin polarization of tunneling electrons across an Al_2O_3 barrier, for which all 3d-ferromagnets show positive SPs [6]. Also recent experiments by LeClair *et al*. [10, 11, 12] demonstrated the decisive role of the electronic structure of the interfaces in SDT.

It is evident, therefore, that the tunneling SP is *not* an intrinsic property of the ferromagnet alone but depends on the structural and electronic properties of the entire junction including the insulator and the ferromagnet/insulator interfaces. This fact makes the quantitative description of transport characteristics of MTJs much more complicated; however, it broadens dramatically the possibilities for altering the properties of MTJs. In particular, by modifying the electronic properties of the tunneling barrier and the ferromagnet/insulator interfaces it is possible to engineer MTJs with properties desirable for device applications.

The main objective of this review article is to address various factors that control the magnitude of magnetoresistance in magnetic tunnel junctions. Starting from early experiments on spin-dependent tunneling and their interpretation, we consider then recent experiments and models, which highlight the role of the electronic structure of the ferromagnets, the insulating layer and the ferromagnet/insulator interfaces. We also discuss the role of disorder in the barrier and in the ferromagnets and their influence on magnetoresistance.

Most recent reviews on spin-dependent tunneling by Levy and Zhang [13] and by Moodera *et al*. [8] appeared in 1999 and summarized experimental and theoretical results published up to this date.

2. Early experiments and models

2.1 Experiments on spin-dependent tunneling

The field of spin-dependent tunneling was founded by the pioneering experiments of Tedrow and Meservey [5, 6]. They used ferromagnet/insulator/superconductor (FM/I/S) tunnel junctions to measure the spin polarization of the tunneling current originating from various ferromagnetic metals across an alumina insulating barrier. In these experiments, electrons tunnel through the barrier to a superconducting Al film which acts as a spin detector. The superconducting density of states (DOS) has a gap of 2Δ in the quasiparticle spectrum and characteristic singularities at $E = \pm\Delta$. If the thin superconducting film (a few nm or less) is placed in a magnetic field **H** applied parallel to the film plane, the quasiparticle states in the superconductor split due to the Zeeman interaction of the magnetic field with the electron spin magnetic moment. In this case, the DOS of the superconductor is the superposition of the up-spin and down-spin contributions separated by energy of $2\mu_B H$, as shown in Fig.2a. The orientation of the magnetic moment and therefore the spin directions are defined by the applied field.

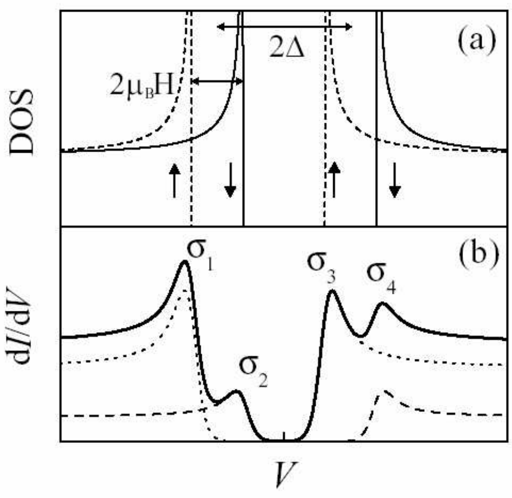

Fig.2 Tunneling in a ferromagnet/insulator/superconductor junction. (a) Density of states of the a superconductor splitted by a value of $2\mu_B H$ into the up- and down-spin contributions. (b) Conductance as a function of voltage for each spin orientation (dotted and dashed lines) and the total conductance (solid line). After Meservey and Tedrow [6].

The sharply peaked DOS of the superconductor makes it possible to separate the contributions from the up- and down-spin electrons in the tunneling current. As a result, tunneling from a ferromagnetic metal into such a superconductor gives rise to an asymmetric conductance curve, which is schematically shown in Fig.2b. This asymmetry is the consequence of the fact that electronic states in the ferromagnetic metal are exchange-split which leads to an unequal DOS in the ferromagnet at the Fermi energy, $\rho^{\uparrow} \neq \rho^{\downarrow}$. Since the $\rho^{\uparrow}$ and $\rho^{\downarrow}$ determine the number of electrons which can tunnel within each spin channel, the spin conductance is weighted with the respective spin DOS. Assuming that spin does not change in the tunneling process, i.e. the total conductance is the sum over the up- and down-spin channels, $G = G^{\uparrow} + G^{\downarrow}$, the tunneling spin polarization can be obtained by measuring the relative heights of the conductance peaks displayed in Fig.1,

$$P = \frac{G^{\uparrow} - G^{\downarrow}}{G^{\uparrow} + G^{\downarrow}} = \frac{(\sigma_4 - \sigma_2) - (\sigma_1 - \sigma_3)}{(\sigma_4 - \sigma_2) + (\sigma_1 - \sigma_3)}. \quad (1)$$

A more accurate determination of the tunneling spin polarization in FM/I/S junctions must account for spin-orbit scattering in the superconductor [6, 8]. Table 1 shows the experimental values of the spin polarization of the tunneling current across Al_2O_3 into superconducting Al from various ferromagnetic 3d-metals corrected for the spin-orbit scattering. Along with the values of *P* obtained in early experiments [6], recently measured values are shown in Table 1. These new values of the spin polarization are higher than the old ones due to improved deposition techniques resulting in cleaner junctions with better interfaces.

Table 1. Tunneling spin polarizations obtained in experiments on FM/Al_2O_3/Al tunnel junctions.

FM	Ni	Co	Fe	$Ni_{80}Fe_{20}$	$Ni_{40}Fe_{60}$	$Co_{50}Fe_{50}$	$Co_{84}Fe_{16}$
P(%), old values [6]	23	35	40	32	–	–	–
P(%), new values [8,14]	33	42	45	48	55	55	55

We note here that although the relatively recent technique of Andreev reflection [15] is also capable of measuring spin polarization, its relevance for magnetic tunnel junctions and TMR values is questionable at best. Andreev reflection weights the contribution of different electronic states differently than tunneling, and further, the influence of the insulating barrier implicit in the tunneling process is not present. Thus, although there does seem to be a rough correspondence between spin-dependent tunneling across Al_2O_3 and Andreev reflection measurements, this correspondence is most likely spurious.

The results of these early experiments on SDT were interpreted in terms of the DOS of the ferromagnetic electrodes at the Fermi energy. Assuming that the spin conductance is proportional to $\rho^{\uparrow}$ for the majority-spin electrons and is proportional to $\rho^{\downarrow}$ for the minority-spin electrons, we arrive at the result that the measured values of the SP of the tunneling conductance, *P*, should be equal to the SP of the DOS at the Fermi energy of the ferromagnet,

$$P_{FM} = \frac{\rho^{\uparrow} - \rho^{\downarrow}}{\rho^{\uparrow} + \rho^{\downarrow}}. \quad (2)$$

This result demonstrates, however, inconsistency between the measured and predicted values of the SP. Indeed, as is evident from Table 1, the SP of the tunneling conductance from all the 3d ferromagnetic metals and their alloys appears to be positive, which implies that the majority-spin electrons tunnel more efficiently than the minority-spin electrons. This is in the contradiction with the bulk band structure, at least, for the two ferromagnetic metals, Co and Ni, which have the dominant contribution of the minority spins at the Fermi energy making the respective SP of the DOS to be negative.

2.2 Stearns' model

This inconsistency between the experimental and theoretical values of the SP is the consequence of the fact that the tunneling conductance depends not only on the number of electrons at the Fermi energy but also on the tunneling probability, which is different for various electronic states in the ferromagnet. The electronic structure of the 3d ferromagnets is characterized by dispersive s bands, which are hybridized with more localized d bands. The latter have a strong weight at the Fermi energy for the minority-spin electrons in Co and Ni that leads to the negative SP of the DOS in these ferromagnets. These features of the electronic band structure were taken into account by Stearns [7], who developed a simple quantitative model to treat the spin polarization of electrons tunneling from various ferromagnetic metals. Stearns pointed out that the transmission probability depends on the effective mass which is different for different bands. The localized d electrons have a large effective mass and therefore decay very rapidly into the barrier region, whereas the dispersive s-like electrons decay slowly. According to this argument, the nearly free-electron, most dispersive bands should provide essentially all the tunneling current. [16]

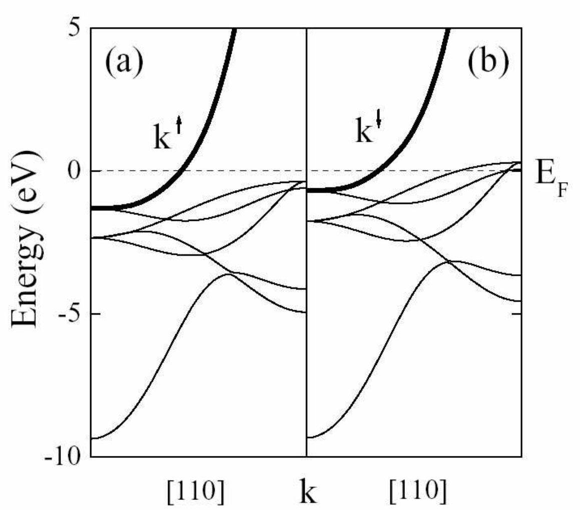

Fig.3 Electronic bands in bulk fcc Ni in [110] direction for the majority- (a) and minority- (b) spin electrons. The heavy lines show the free-electron-like bands which dominate tunneling. $k^{\uparrow}$ and $k^{\downarrow}$ are the Fermi wave vectors which determine the spin polarization of the tunneling current. After Stearns [7].

Stearns' idea is illustrated in Fig.3, which shows the electronic bands of bulk Ni in the [110] direction. As is seen from this figure, for the majority-spin electrons the dispersive s band is the only one that crosses the Fermi energy (Fig. 3a). For the minority spins, however, there are several bands that appear at the Fermi level (Fig. 3b). According to Stearns' argument, the dispersive band that is indicated in the Fig. 3b by the heavy solid line is the only band contributing to the tunneling process – the other localized bands are to be essentially disregarded.

The dispersive bands that dominate tunneling are similar to free-electron bands, and therefore, the DOS of these bands at the Fermi energy is proportional to their Fermi wave vector. Assuming that the conductance is proportional to the DOS of these itinerant electrons we can rewrite Eq.(2) for the SP of the ferromagnet as

$$P_{FM} = \frac{k^{\uparrow} - k^{\downarrow}}{k^{\uparrow} + k^{\downarrow}}, \quad (3)$$

where $k^{\uparrow}$ and $k^{\downarrow}$ are the Fermi wave vectors of the dispersive bands for the majority and minority spins. Using an accurate analysis of the electronic band structure, Stearns found that P_{FM}=45% for Fe and P_{FM} =10% for Ni which are consistent with the experimental data.

Stearns actually introduced a notion of "tunneling density of states". This notion was used by other researchers (e.g., ref. [17]) to designate the effective number of electrons which can tunnel from one ferromagnetic metal and the number of effective empty states available in the other ferromagnetic metal, so that the tunneling conductance per spin is proportional to their product. In the model proposed by Stearns, the tunneling DOS is identified as the Fermi wave vectors of the itinerant electrons with corresponding spin. The results obtained by Stearns are an early indication of the fact that the understanding of spin-dependent tunneling requires a detailed knowledge of the electronic structure of MTJs.

2.3 Julliere's experiments and model

An important advance was made by Julliere [1] in 1975, a few years later after the successful experiments on SDT were reported. In these experiments the superconducting film was replaced by another ferromagnetic metal film, thereby making a FM/I/FM tunnel junction. It was reasoned that instead of using magnetic-field-induced spin-split states of a superconductor as a spin detector it is possible to use exchange-split states of another ferromagnet. In this case, it was expected that the tunneling current should depend on the relative magnetization orientation of the two ferromagnetic electrodes, giving rise to TMR. This is in fact what was observed. Using Co and Fe ferromagnetic films with different coercive fields and a Ge barrier layer Julliere observed sizable magnetoresistance at 4.2K. The maximum TMR value was found about 14% at zero bias, but decreased very rapidly with increasing bias voltage, as shown in Fig.4. This rapid decrease in TMR was attributed to spin-flip scattering at ferromagnet/barrier interfaces.

Julliere interpreted these results in terms of a simple model, which is based on two assumptions. First, he assumed that spin of electrons is conserved in the tunneling process. It follows, then, that tunneling of up-spin and down-spin electrons are two independent processes, so that the conductance occurs in the two independent spin channels. Such a two-current model is also used to interpret the closely related phenomenon of giant magnetoresistance (see, e.g., [18]). According to this assumption, electrons originating from one spin state of the first ferromagnetic film are accepted by unfilled states of the same spin of the second film. If the two ferromagnetic films are magnetized parallel, the minority spins tunnel to the minority states and the majority spins tunnel to the majority states. If, however, the two films magnetized antiparallel the identity of the majority- and minority-spin electrons is reversed so that the majority spins of the first film tunnel to the minority states in the second film and vice versa.

Second, Julliere assumed that the conductance for a particular spin orientation is proportional to the product of the effective (tunneling) density of states of the two ferromagnetic electrodes. According to these assumptions, the conductance for the parallel and antiparallel alignment, G_P and G_{AP}, can be written as follows:

$$G_P \propto \rho_1^{\uparrow}\rho_2^{\uparrow} + \rho_1^{\downarrow}\rho_2^{\downarrow}, \quad (4)$$

$$G_{AP} \propto \rho_1^{\uparrow}\rho_2^{\downarrow} + \rho_1^{\downarrow}\rho_2^{\uparrow}, \quad (5)$$

where $\rho_i^{\uparrow}$ and $\rho_i^{\downarrow}$ are the tunneling DOS of the ferromagnetic electrodes (designated by index i = 1, 2) for the majority- and minority-spin electrons. It follows from Eqs. (4) and (5) that the parallel- and antiparallel-magnetized MTJ have a different conductance, which implies a non-zero TMR. We define TMR (following the majority of researchers) as the conductance difference between parallel and antiparallel magnetizations, normalized by the antiparallel conductance, i.e.

$$TMR \equiv \frac{G_P - G_{AP}}{G_{AP}} = \frac{R_{AP} - R_P}{R_P}. \quad (6)$$

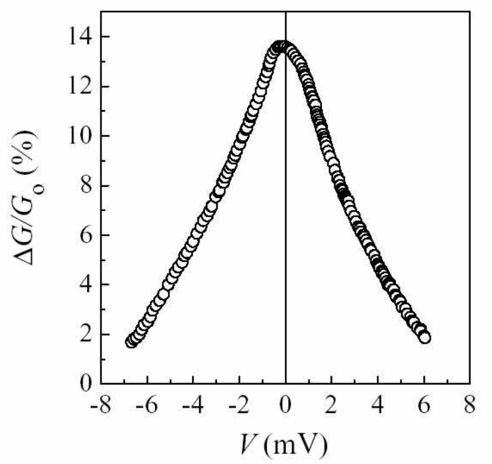

Fig.4 Original demonstration of the tunneling magnetoresistance effect. The relative conductance change due to an applied magnetic field versus applied bias in a Fe/Ge/Co junction at 4.2 K. After Julliere [1].

Using Eqs. (4) and (5), we arrive then at Julliere's formula

$$TMR = \frac{2P_1P_2}{1 - P_1P_2}, \quad (7)$$

which expresses the TMR in terms of the effective SPs of the two ferromagnetic electrodes

$$P_i = \frac{\rho_i^{\uparrow} - \rho_i^{\downarrow}}{\rho_i^{\uparrow} + \rho_i^{\downarrow}}, \quad (8)$$

where i = 1, 2. Using the known values of the spin polarization for Co and Fe from experiments of Tedrow and Meservey [5], Julliere deduced the TMR value of 26%, which is larger than the maximum measured value of 14%. He explained the discrepancy by magnetic coupling between the ferromagnetic electrodes and spin-flip scattering.

These results of Julliere stimulated further research in the field of magnetic tunnel junctions. Unfortunately, they were not reproduced by other researchers, and their true interpretation is still the subject of debate. Nevertheless, the importance of the paper by Julliere should not be underestimated, in particular, his simple quantitative model which was later used by many researchers to correlate the magnitude of TMR in MTJs with the SP of ferromagnets measured in experiments on FM/I/S tunnel junctions.

2.4 Slonczewski's model

First accurate theoretical consideration of TMR was made by Slonczewski in 1989 [19]. He considered tunneling between two identical ferromagnetic electrodes separated by a rectangular potential barrier assuming that the ferromagnets can be described by two parabolic bands shifted rigidly with respect to one another to model the exchange splitting of the spin bands. Having imposed perfect translational symmetry of the tunnel junction along the layers and matched the wave functions of electrons across the junction, he solved the Schrödinger equation and determined the conductance as a function of the relative magnetization alignment of the two ferromagnetic films. In the limit of thick barrier, he found that the conductance is a linear function of the cosine of angle Θ between the magnetic moments of the films,

$$G(\Theta) = G_0 \left(1 + P^2 \cos\Theta\right). \tag{9}$$

Here P is the effective spin polarization of tunneling electrons given by

$$P = \frac{k^{\uparrow} - k^{\downarrow}}{k^{\uparrow} + k^{\downarrow}} \frac{\kappa^2 - k^{\uparrow}k^{\downarrow}}{\kappa^2 + k^{\uparrow}k^{\downarrow}}, \tag{10}$$

where κ is the decay constant of the wave function into the barrier which is determined by the potential barrier height U, $\kappa = \sqrt{(2m/\hbar^2)(U - E_F)}$. As follows from Eq.(10), in addition to the factor that represents the spin polarization P_{FM} of the ferromagnet (3) the SP of the tunneling current contains a factor which depends on the barrier height. In the limit of a high barrier it tends to unity reducing Slonczewski's result for TMR to Julliere's formula. However, if the barrier is not very high and the decay constant is comparable to or less than the wave vectors of electrons in the ferromagnetic metals, the magnitude of TMR decreases with decreasing U and even changes sign for sufficiently low barriers, which is demonstrated in Fig.5.

Slonczewski wrote that these "results contradict the plausible notion that the spin polarization P is characteristic of the electron structure of the electrode *alone* and would have the same value (sign at least) in any tunneling experiment." This was the fist important indication of the fact that the SP of the conductance is not intrinsic property of the ferromagnets.

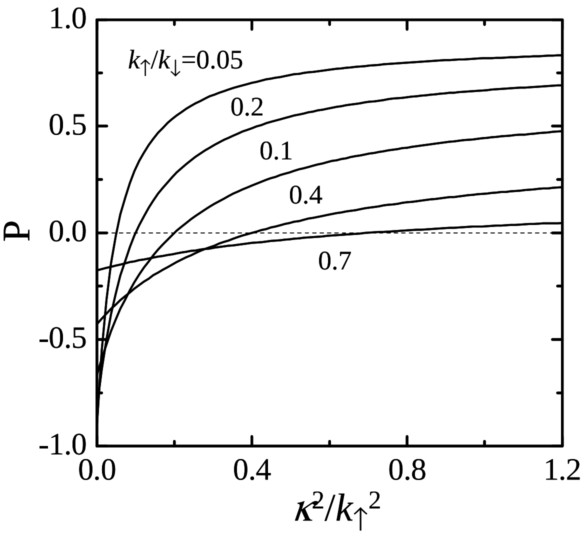

Fig.5 Spin polarization of the tunneling conductance as a function of the normalized potential barrier height for various values of $k_{\uparrow}/k_{\downarrow}$. After Slonczewski [19].

3. Recent experiments

Over the next two decades, several groups attempted to perform experiments on magnetic tunnel junctions using different ferromagnetic electrodes and barrier layers (e.g. [17, 20, 21, 22, 23, 24]). In all cases, the observed TMR values were at most a few percent at relatively low temperatures. In particular, Maekawa and Gäfvert [17] used Ni/NiO/Co junctions to study TMR and to correlate it with the magnetization loops of the ferromagnetic electrodes. They found TMR values of about 2% at 4.2K, which rapidly decreased with increasing temperature. Although these values were much less than those anticipated, this work clearly indicated that the conductance variation as a function of applied magnetic field was indeed due to the change in the relative magnetization alignment of the two ferromagnetic films.

Only in 1995, nearly 20 years after the first experiments on TMR, Miyazaki and Tezuka [2] and Moodera *et al*. [3] independently demonstrated >10% TMR at room temperature. In the both experiments MTJs based on alumina as a barrier layer separating transition-metal electrodes were used. The results of Moodera *et al*. for the resistance change in a CoFe/Al_2O_3/Co MTJ as a function of applied magnetic field are shown in Fig.1. The latter experiments demonstrated the fabrication procedure which provides MTJs with a pinhole-free Al_2O_3 tunnel barrier and with smooth interfaces resulting in reproducible, high TMR values at room temperature. These achievements quickly garnered a great deal of attention, and catalyzed many groups to investigate MTJs. In the next few sections, we discuss most important features of TMR observed experimentally, in particular, the dependence of TMR on magnetic field, bias voltage and temperature, on the type of the ferromagnet and its crystallographic orientation, on the barrier material and interface properties.

3.1 Magnetic field dependence

In order to observe the TMR phenomenon one needs to realize experimentally both parallel (P) and antiparallel (AP) magnetization alignment in a MTJ. Perhaps the simplest way to provide this is to use two ferromagnetic layers with different coercive fields, for example, hard and soft ferromagnets such as Co (hard) and $Ni_{80}Fe_{20}$ (soft). The typical behavior of TMR versus magnetic field is shown in Fig.6a for a $Ni_{80}Fe_{20}/Al_2O_3/Co$ junction. When the field is swept through zero and reaches values between the $Ni_{80}Fe_{20}$ and Co coercive fields, an antiparallel magnetization alignment is reached between $\pm 0.5 - 1.5$ mT.

Exchange biasing is another way to realize the P and AP magnetization alignment. In this case, one of the magnetic electrodes is in direct contact with an antiferromagnetic (AFM) material (e.g., FeMn or NiO). The presence of an exchange anisotropy at the FM/AFM interface shifts the entire magnetization loop away from zero field, such that it is centered at a finite magnetic field [25]. Typical TMR behavior for an exchange-biased system is displayed in Fig. 6b. Technologically, exchange biasing is advantageous because the resistance transition takes place near zero magnetic field, and it generally results in greater magnetic stability, which is important for technological applications of MTJs [26, 27].

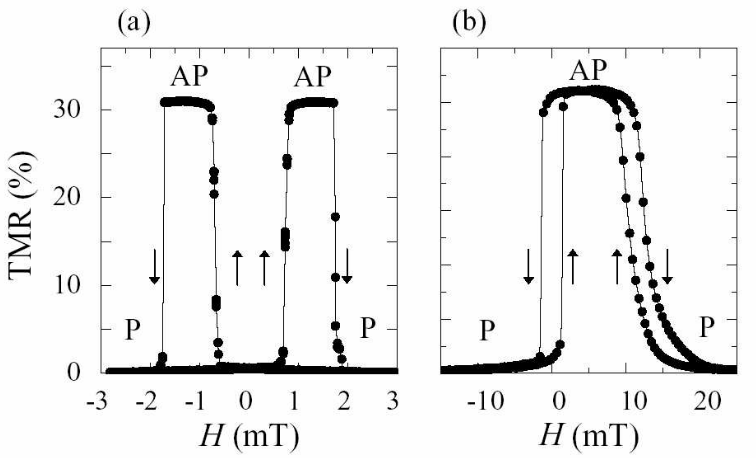

Fig.6 Magnetoresistance versus magnetic field for a hard-soft MTJ (a) and an exchange-biased MTJ (b), both at 10 K. Vertical arrows refer to sweep direction. Both curves are taken at $V = 0$. From LeClair [28].

According to Slonczewski's model (Sec.3.4) a magnetic tunnel junction should work as a spin polarizer of the electric current if the magnetization of the one ferromagnetic film rotates with respect to the magnetization of the other. Indeed, the predicted cosine variation of the TMR was found in the experiments by Moodera and Kinder [29]. Using electrodes with different coercive fields they measured the angular dependence of magnetoresistance, which is shown in Fig.7. At a field higher than the coercive field of one electrode, the magnetization of the softer film follows the field direction, when the sample is rotated. This changes gradually the relative magnetization orientation of the two ferromagnetic films from parallel to antiparallel. It was found that the resistance variation follows the cos Θ dependence, thereby supporting the FM/I/FM tunneling model [30]. When similar measurements were done at a field value higher than the coercive field of the both electrodes no resistance change was found.

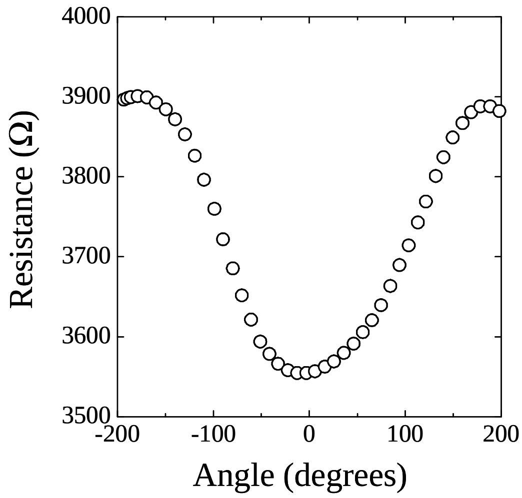

Fig.7 Angular dependence of the magnetoresistance of a $CoFe/Al_2O_3/Co$ junction measured in a magnetic field lower than the coercive field of one electrode but higher than the coercive field of the other electrode. After Moodera and Kinder [29].

3.2 Voltage Dependence

In most magnetic tunnel junctions the magnitude of TMR decreases strongly with increasing bias voltage, similar to that observed originally by Julliere (see Fig.4). A figure of merit is the voltage at which the TMR is reduced by a factor of two. In the work of Julliere, only 3mV bias was needed to halve the TMR value (Fig.4). Later Moodera *et al*. fabricated junctions with a "half voltage" of 200mV [3]. With improving control of the barriers, several groups increased this value up to more than 500mV (e.g., [31, 32]).

In order to explain this drop in TMR with bias, Zhang *et al*. [33] proposed a model suggesting that inelastic scattering by magnon excitations at the ferromagnet/insulator interface control the voltage dependence. In the presence of non-zero bias, electrons which have tunneled across the barrier arrive at the second ferromagnet as hot electrons with energy higher than the Fermi energy of this electrode (provided that no inelastic scattering event has occurred). These hot electrons may then lose their energy by emitting a magnon and thereby flipping the electron spin. With increasing bias voltage more magnons can be emitted resulting in the reduced TMR values. By using one parameter, in addition to parameters that fix the response at zero bias, Zhang *et al*. explained the softening of the TMR with bias up to 200 mV. Later, this model was used by Han et al. [34], who have performed a careful analysis of the conductance and magnetoresistance as a function of voltage and temperature for $Co_{75}Fe_{25}/Al_2O_3/Co_{75}Fe_{25}$ tunnel junctions.

Although these results and also experiments by Moodera *et al*. [35] suggest that the bias dependence of TMR is due to magnon excitations at the interface, recent experiments by Wulfhekel *et al*. [36] seem to be inconsistent with this plausible explanation. Using spin-polarized tunneling microscopy on Co (0001) they studied TMR in a "MTJ" in which vacuum served as a barrier separating a ferromagnetic STM tip and the Co electrode. Contrary to oxide barriers used in normal MTJs, the barrier in these experiments was "perfect" and did not suffer from any imperfections of the oxide. On the other hand, magnon excitations at the interfaces (surfaces of the ferromagnets) could be present. Wulfhekel *et al*. found, in sharp contrast to the FM/Al_2O_3/FM junctions, no variation in TMR up to ±0.9V at relatively large separations between the ferromagnetic tip and the Co surface. They concluded that most of the voltage dependence is not related to magnon excitations at the interface and put forward a model of Zhang and White [37] who had suggested that the voltage drop in TMR is due to localized trap states in the amorphous barrier. The impurity-assisted contribution to the bias dependence of TMR is also supported by experiments of Jansen and Moodera [38]. Interestingly, the latter mechanism can even result in an *increased* TMR relative to pure junctions, as demonstrated by Jansen and Moodera for Fe-doped NiFe/Al_2O_3/CoFe junctions [39].

Another mechanism, which could contribute to the voltage dependence of the conductance and TMR, is related to the electronic structure of the ferromagnets. Biasing a MTJ leads to the contribution from electrons which tunnel from the occupied states below the Fermi energy of the one electrode to the empty states above the Fermi energy of the other electrode. Due to the change in the electronic structure of the ferromagnets (e.g., the DOS) as a function of energy, the conductance should be energy-dependent resulting in the variation of TMR versus the applied voltage. This mechanism should obviously be sensitive to the type of ferromagnets. Surprisingly, however, for the alumina-based MTJs with different ferromagnetic electrodes the band structure effects in the voltage dependence have not been reported until recently. Only recently, using Co/Al_2O_3/Co MTJs with fcc- and hcp-Co electrodes, LeClair *et al*. [40] found a relationship between the magnetotransport properties and the calculated DOS for the two different crystalline phases of Co. The influence of the electronic structure on the voltage dependence of TMR for MTJs with insulators different from alumina was found in refs. [9, 41] and will be discussed in Sec.3.5.

3.3 Temperature dependence

In all tunnel junctions the TMR decreases with increasing temperature. As was first noticed by Shang *et al*. [42], the temperature dependence of the tunnel resistance for MTJs greatly exceeds that for nonmagnetic junctions with nominally identical barriers. Typically, Al/Al_2O_3/Al junctions showed only a 5-10% change in resistance from 4.2 to 300 K, while MTJs always exhibited a 15-25% change in resistance, as shown in Fig.8 for a Co/Al_2O_3/Co junction. The TMR can decrease as much as 25% or more from 4.2 to 300 K depending on the magnetic electrodes. Shang *et al*. explained these results within a simple phenomenological model, in which they assumed that the tunneling spin polarization P decreases with increasing temperature due to spin-wave excitations, as does the surface magnetization. They thus assumed that both the tunneling spin polarization and the interface magnetization followed the same temperature dependence, the well-known Bloch $T^{3/2}$ law, $M(T) = M(0)(1 - \alpha T^{3/2})$. By fitting parameter α Shang *et al*. obtained a satisfactory explanation for the temperature dependence of TMR, as demonstrated by the fit in Fig.8a. MacDonald *et al*. [43] provided a more rigorous theoretical justification of these ideas, essentially reproducing the proportionality between $M(T)$ and P(T).

Another mechanism which can cause the reduction of TMR with temperature is spin-flip scattering by magnetic impurities in the barrier. As was shown by Vedyayev *et al*. [44], the number of electrons contributing to this process increases with increasing temperature resulting in the drop of TMR. In addition, inelastic scattering which does *not* flip the spin, such as electron-phonon scattering, can also cause the reduction of TMR in the presence of localized states in the barrier. This was recently demonstrated by Tsymbal *et al*. [45], who considered spin-dependent transport across an amorphous barrier, and showed that spin-conserving inelastic scattering is detrimental to TMR.

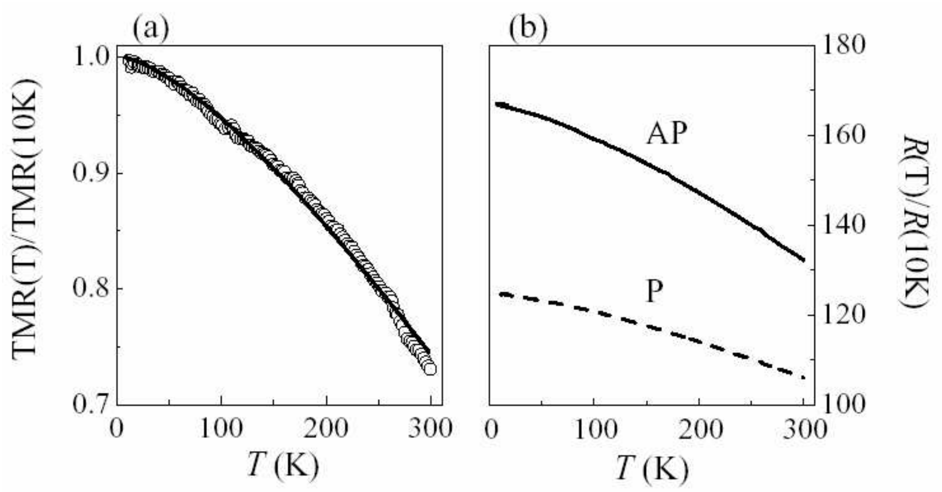

Fig.8 (a) Temperature dependence of TMR for a Co/Al_2O_3/Co MTJ (circles) along with a fit to the model of Shang *et al*. [42] (line). (b) Resistance versus temperature for parallel (P, dashed line) and antiparallel (AP, solid line) magnetization alignment for the same junction. From LeClair [28].

3.4 Ferromagnet dependence

In general, the most recent spin polarization values with Al_2O_3 barriers obtained via the SDT technique [8, 14] agree well with the maximum TMR values reported with Al_2O_3 barriers [8, 46] within Julliere's model. Table 2 compares the expected TMR values based on Julliere's model, using values of SP obtained from SDT experiments, with measured TMR values using the *same* barriers in both cases. However, we caution that Julliere's model is only a phenomenological guide to estimate the magnitude of the TMR effect when tunneling spin polarizations are known.

Table 2. Comparison of TMR values expected from Julliere's model (using SP data obtained via the Meservey-Tedrow technique) with measured low-temperature TMR values, in each case using the same tunnel barrier.

Junction	TMR (%)		Ref.
	Julliere	Expt.	
Ni/Al_2O_3/Ni	25	23	28
Co/Al_2O_3/Co	42	37	28
$Co_{75}Fe_{25}$/Al_2O_3/$Co_{75}Fe_{25}$	67-74	69	46
LSMO/$SrTiO_3$/LSMO	310	1800	47

Obviously, one expects the largest TMR values for materials with the largest tunneling spin polarization. This explains a great deal of the recent interest in so-called “half-metallic” ferromagnets, materials for which only one spin band is occupied at the Fermi level, resulting in perfect 100% spin polarization [48]. Many compounds have been predicted to be half metallic, such as the half- and full-Heusler alloys NiMnSb [49] and Co_2MnSi [50]; the oxides CrO_2 [51], Fe_3O_4 [52, 53], and $La_{0.67}Sr_{0.33}MnO_3$ [54, 55]; and the sulfide $Co_xFe_{1-x}S_2$ [56]. However, only $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) [57], NiMnSb [58], and CrO_2 [59, 60] have any experimental evidence in favor of half metallic behavior. LSMO has been successfully used as electrodes in MTJs by Lu *et al*. [61] and Viret *et al*. [62], who observed TMR effects of more than 400% at low temperature utilizing $SrTiO_3$, $PrBaCu_{2.8}Ga_{0.2}O_7$, or CeO_2 barriers. Using Eq.(7), this implies a spin polarization of more than 80%, in agreement with SDT experiments [63]. Sun [64] has also very recently reported more than 100% TMR as well in LSMO/$SrTiO_3$/LSMO junctions. Similarly, Jo *et al*. [65] have used another mixed-valence manganite, $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) and investigated LCMO/$NdGaO_3$/LCMO and LCMO/$NdGaO_3$/LSMO MTJs, also observing more than 400% TMR. More recently, Bowen *et al*. [47] have observed 1800% TMR in LSMO/$SrTiO_3$/LSMO junctions, implying a spin polarization of 95% based on Julliere’s model and essentially corroborating photoemission results showing LSMO to be half-metallic. An extensive review of magnetotransport phenomenon in magnetic oxides can be found in [66].

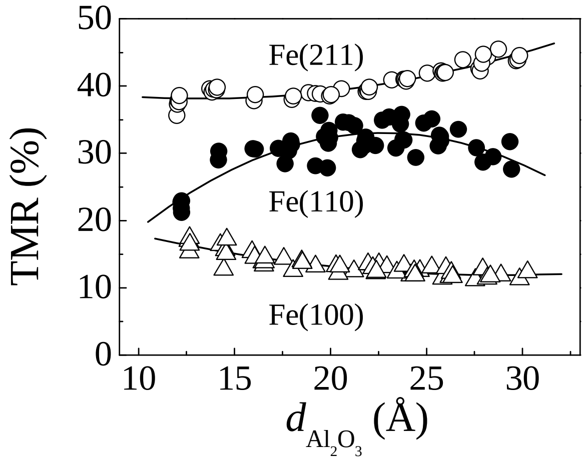

Fig.9 TMR at 2K as a function of Al_2O_3 thickness for Fe(211), Fe(110), and Fe(100) epitaxial electrodes in Fe/Al_2O_3/CoFe junctions. Lines are only a guide to the eye. From Yuasa *et al*. [67].

Given the expected dependence of TMR on the electronic structure of ferromagnetic electrodes, one would anticipate a dependence on the crystallographic orientation of the electrodes. However, MTJs with even a single epitaxial layer are notoriously difficult to fabricate, and only recently have semi-epitaxial (i.e., one epitaxial electrode) MTJs been grown. Yuasa *et al*. [67] have prepared Fe(100,110,211)/Al_2O_3/CoFe MTJs with the epitaxial bottom Fe layer to study the effect of the Fermi surface anisotropy on transport properties. They observed a strong dependence of the TMR on crystallographic orientation, as shown in Fig.9. This fact clearly points to the details of the Fe band structure [68] and momentum filtering. Naively looking at the most dispersive *s*-like bands near E_F in Fe [68], the trend that TMR[Fe(211)] >

TMR[Fe(110)] > TMR[Fe(100)] can perhaps be justified in some way, though, as we will see in Sec.4.3, it is expected that the Fe(100) tunneling spin polarization should be much larger. Another possible reason for the dependence of the TMR on crystallographic orientation could be a slightly different growth mode of the amorphous Al_2O_3 on the different crystalline facets of Fe, giving rise to a slightly different barrier quality for each electrode orientation and barrier thickness. However, there is no direct evidence for this mechanism.

3.5 Barrier dependence

Work to date on MTJs has focused almost exclusively on Al_2O_3 tunnel barriers, for a variety of reasons. Perhaps most important are the ease in fabricating ultrathin, pinhole-free Al_2O_3 layers, spin conservation demonstrated across Al_2O_3 barriers [6], and the first successful demonstrations of the TMR effect used Al_2O_3 barriers [2, 3, 4]. Significant efforts have been invested to characterize, understand and improve properties of alumina barriers (see, e.g., refs. [69, 70, 71, 72, 73, 74]). Nevertheless, in the last several years several alternative barriers have been successfully employed, some with very distinct behavior compared to Al_2O_3.

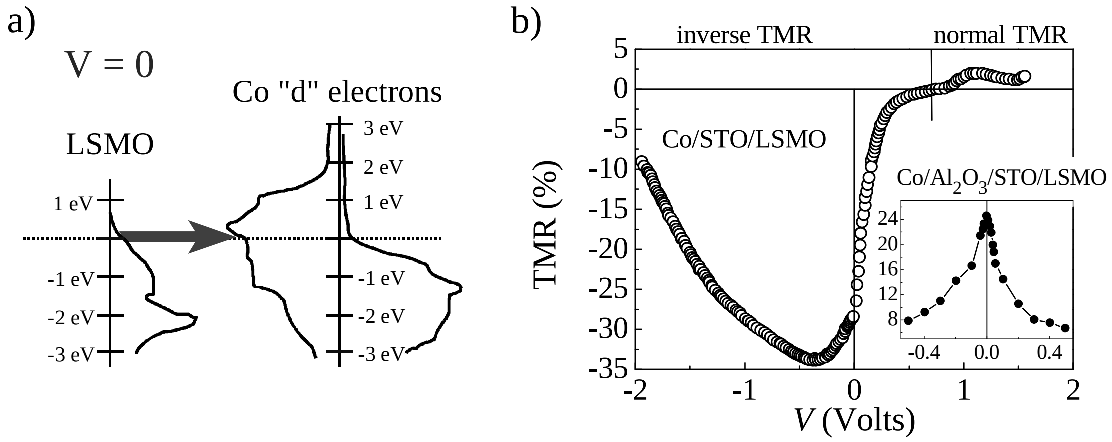

Fig. 10 (a) Schematic of the spin-polarized densities of states of LSMO (as derived from photoemission) and the Co(100) surface (calculated). (b) TMR ratio versus applied bias for a Co/$SrTiO_3$/LSMO junction at 5K. Inverse TMR is observed for V < 0.8 Volts, while normal TMR is observed for V > 0.8 Volts, indicating that the Co/$SrTiO_3$ spin polarization is negative for V < 0.8 Volts. Inset: TMR ratio versus applied bias for a Co/Al_2O_3/$SrTiO_3$/LSMO junction. In this case, the polarization of Co/Al_2O_3 and LSMO are both positive, and a normal (positive) TMR is seen. From de Teresa *et al*. [9].

Probably the most remarkable result was obtained by de Teresa *et al*. [9], who found that the tunneling spin polarization depends explicitly on the insulating barrier. They used half metallic LSMO as one of the electrodes with Al_2O_3 or $SrTiO_3$ barriers, or with a composite Al_2O_3/$SrTiO_3$ barrier. Since it is known that LSMO has only majority states at E_F, the tunneling spin polarization must be positive and close to 100% [63, 57], regardless of the insulating barrier used. As expected, de Teresa *et al*. found that Co/Al_2O_3/LSMO MTJs have a positive TMR for all biases. Surprisingly, however, Co/$SrTiO_3$/LSMO junctions showed *negative* TMR values at zero bias, as shown in Fig.10b. Teresa *et al*. explained these results in terms of the SP of ferromagnet-barrier interfaces rather than the SP of ferromagnets alone. They proposed that the polarization of the Co/$SrTiO_3$ interface must be *negative*, opposite that of Co/Al_2O_3 interfaces. In order to show this more conclusively, they investigated Co/Al_2O_3/$SrTiO_3$/LSMO junctions, with the expectation that since the LSMO and Co/Al_2O_3 tunneling spin polarizations are both positive, a normal positive TMR would result for all biases. As shown in the inset to Fig. 10b, a normal positive TMR is indeed observed for all biases, with a bias dependence that is essentially identical to standard Co/Al_2O_3/Co junctions. Teresa *et al*. [9] interpreted the sign change of the Co tunneling spin polarization in terms of interface bonding, the effect of which was proposed earlier by Tsymbal and Pettifor [75] and will be discussed in Sect. 4.2.

More recent results by Sugiyama *et al*. [76] and Sun *et al*. [77] essentially corroborated these results. Experiments by Sharma *et al*. [41] utilizing Ta_2O_5/Al_2O_3 composite barriers showed that the sign of the spin polarization at Ta_2O_5 interfaces varies with bias voltage and proposed an explanation similar to that of de Teresa *et al*. [9]. All these results clearly illustrate the rich physics behind spin-dependent tunneling in MTJs, as well as the intriguing possibility of "engineering" MTJs with tailored properties.

Among other barriers MgO has been used by a number of researchers. Platt *at al*. [78] first demonstrated a large TMR (about 20% at 77 K) in MTJs based on a reactively sputtered MgO barrier. Recently a few successful attempts have been made to grow *epitaxial* MgO barriers. Wulfhekel *et al*. [79] and Klaua *et al*. [80] fabricated epitaxial Fe/MgO/Fe MTJs on Fe whiskers and extensively investigated the growth and the local transport properties of these junctions. TMR studies of these MTJs were, however, hampered by the difficulties in decoupling the magnetic electrodes from the Fe whisker substrate. Successful magnetotransport experiments on epitaxial Fe/MgO/FeCo(100) MTJs have recently been performed by Bowen *et al*. [81] who demonstrated 27% TMR at room temperature, which increased to 60% at 30 K. From the bias dependence of TMR and the theoretical predictions [82], they concluded that *s*-character electrons are predominantly tunneling across 20Å MgO barrier. Among other barriers HfO_2 [83] and Ta_2O_5 [78, 84] were successfully used in magnetic tunnel junctions.

MRAM and sensor applications of MTJs require, in addition to high values of TMR, a reduced resistance-area product (RA). Typical values which are required are 100 $\Omega\cdot\mu m^2$ for MRAMs and less than 0.5 $\Omega\cdot\mu m^2$ with TMR at least 10 % for filed sensors. These requirements stimulated research on various oxide barriers. Sharma *et al*. [85] have fabricated AlN and AlO_xN_y barriers, observing up to 18% TMR at room temperature with a lower RA product than similar alumina-based junctions as well as a less severe voltage dependence of the TMR. Similarly, Wang *et al*. [86] have fabricated junctions with AlO_xN_y with as little as <10% O present, and found TMR values ranging from 13 to 33% and RA products from 73-8500 $\Omega\cdot\mu m^2$, comparable to pure Al_2O_3. Li *et al*. [87] has reported similar results with Ga_2O_3. Freitas *et al*. have demonstrated MTJs with ZrO_2 [88] or $ZrAlO_x$ [89] barriers, and in both cases the TMR values were comparable to similar junctions with Al_2O_3 barriers but with a much reduced RA product. Thus, at the present time, ZrO_2, $ZrAlO_x$, AlN, AlO_xN_y, and Ga_2O_3 barrier can be considered as alternatives to Al_2O_3 for memory and sensor applications of MTJs.

Up until now, we have focused on tunneling between ferromagnetic electrodes which served as the source of spin polarization. However, the tunneling spin polarization can be obtained (even with nonmagnetic electrodes) due to a spin-dependent tunneling probability. The latter may be achieved by utilizing a magnetic tunnel barrier, such as EuS, which is a ferromagnetic semiconductor with T_C=16.7K [90, 91]. Below T_C the EuS conduction band is exchange split, and tunneling electrons see a spin-dependent barrier height. For EuS, a typical barrier height is about 2eV [92], with a conduction band exchange splitting of ~0.36eV [90, 91]. Given the exponential dependence of tunnel current on barrier height, a highly spin-polarized current is expected.

The principle of spin filtering has been experimentally demonstrated in field emission [91, 93] and in SDT [92] experiments. In the latter case, Moodera *et al*. [92] performed SDT experiments using an Al/EuS/M junctions, where M was Ag, Au, or Al, and found a tunneling spin polarization of approximately 80%. Further, using a related Eu-chalcogenide, EuSe, Moodera *et al*. were able to demonstrate essentially 100% spin polarization. Thus, spin filtering can be considered as an attractive route for the generation and manipulation of highly spin-polarized currents. In particular, by combining ferromagnetic electrodes and spin filtering new hybrid devices and novel effects may be imagined (see, e.g., [94]).

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3.6 Interface dependence

The tunneling current in MTJs is very much influenced by the electronic structure around the interfaces between the ferromagnetic electrodes and the insulating barrier. One of the routes to explore the interface sensitivity is the insertion of ultrathin layers (often called “dusting” layers) at the electrode-barrier interfaces. Tedrow and Meservey [95] used the SDT technique to study the spin polarization of ultrathin ferromagnets and showed that only a few monolayers of ferromagnetic material are needed for full tunneling spin polarization. Moodera *et al*. [96] applied this method to measure the spin polarization in Al/Al_2O_3/Au/Fe junctions as a function of Au interlayer thickness. They found that SP decreased exponentially for the first two monolayers Au, but decreased as 1/*d* at larger thicknesses. In the context of MTJ's, Parkin investigated TMR as a function of the thickness of a nonferromagnetic layer grown on Al_2O_3 [97]. In these experiments, a large tunneling spin polarization was, surprisingly, maintained over distances in excess of 10nm, in striking contrast with the earlier experiments of Moodera [96], as well as later experiments by Sun and Freitas for Cu on Al_2O_3 [98].

Recently LeClair *et al*. [10] have shown that this apparent discrepancy is related to the growth of MTJs. They grew Cu interlayers both above *and* below the Al_2O_3 barrier, which resulted in two different TMR decay lengths, as shown in Fig.11a. For Cu above the Al_2O_3 barrier, the length scale was roughly three times larger than for Cu below the Al_2O_3 barrier. They were able to show that Cu grows on Al_2O_3 in a three-dimensional manner, giving rise to an artificially inflated TMR decay. It was further shown that in Co/Cu/Al_2O_3/Co junctions, where Cu was grown on Co rather than on Al_2O_3, near-ideal Cu growth resulted. In this latter system, LeClair *et al*. found that the normalized TMR (i.e., $TMR(d_{Cu})/TMR(d_{Cu=}0)$) decayed approximately exponentially with increasing Cu thickness, $\exp(-d_{Cu}/\xi)$. Fitting the TMR decay gave ξ~0.26 nm equivalent to just more than one monolayer Cu. Appelbaum and Brinkman [99] first pointed out that tunneling in non-superconducting junctions should be sensitive to the density of states within a few Fermi wavelengths (i.e., $1/k_F$) of the electrode-barrier interface. In this case ξ is about $3.5k_F^{-1}$ suggesting that indeed k_F^{-1} may be the relevant length scale, at least in disordered systems. This is supported by the results of Moodera *et al*. [100] with Ag and Au interlayers, which have almost the same value of k_F^{-1} and give a length scale similar to Cu interlayers, as do Pt interlayers.

A further demonstration of interface sensitivity was subsequently obtained by LeClair *et al*. [11], using ultrathin Cr layers in Co/Cr/Al_2O_3/Co MTJs. The TMR decay was again approximately exponential, and in this case was even more rapid, (see Fig.11b) with ξ~0.1 nm, or only ~0.5 monolayers of Cr. In these experiments, they also added an additional Co layer on top of the Cr dusting layer, i.e., Co/Cr(d_{Cr})/Co/Al_2O_3/Co, shown in Fig.11b. Strikingly, the TMR was almost completely restored with only 3-5 monolayers of Co. This further confirms that only the few monolayers of the electrode adjacent to the ferromagnet-insulator interface dominate MTJ properties, in very good agreement with earlier SDT work on ultrathin magnetic layers.

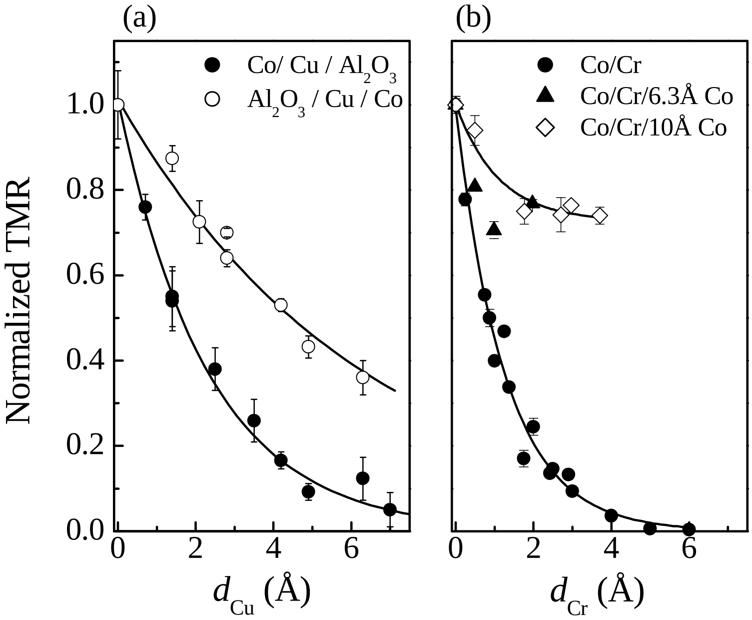

Fig. 11 (a) Normalized TMR at 10K as a function of Cu thickness for Co/Cu(d_{Cu})/Al_2O_3/Co and Co/Al_2O_3/Cu(d_{Cu})/Co. (b) Normalized TMR at 10K as a function of Cr thickness for Co/Cr(d_{Cr})/Al_2O_3/Co and Co/Cr(d_{Cr})/Co(d_{Co})/Al_2O_3/Co tunnel junctions. Adding only a few monolayers of Co on Cr almost completely restores the TMR, demonstrating the interfacial sensitivity of MTJs. Lines are fits to an exponential decay. From LeClair *et al*. [10, 11].

A theory predicts an oscillatory dependence of TMR on interlayer thickness due to quantum-well states formed in the interlayer [101, 102] (see also Sec. 5.1) Although no quantum well states were observed in the studies above, in retrospect one would not expect to observe them except in a nearly-perfect epitaxial system, as is the case for quantum-well states in metallic multilayers. Yuasa *et al*. [103] have recently prepared Co(001)/Cu(001)/Al_2O_3/$Ni_{80}Fe_{20}$ junctions with the bottom epitaxial Co/Cu electrodes and observed true quantum-well oscillations of the TMR that are shown in Fig.12. A clear (damped) oscillation of the TMR is evident, with a period of 11.4Å, in good agreement with the Fermi surface of Cu. Further, independent measurements on similarly grown Co/Cu/Co trilayers gave an oscillation of the interlayer exchange coupling with a period of 11Å. This clear correlation suggests that the oscillation indeed arises from spin-dependent reflection at the Co/Cu interface due to the formation of spin-polarized quantum well states within the Cu interlayer.

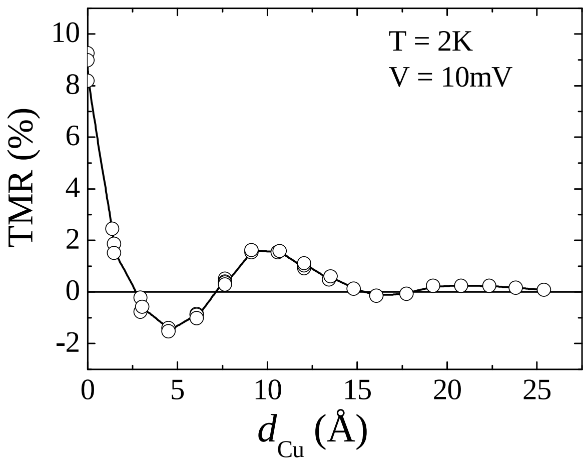

Fig.12 TMR at 2K and a bias of 10mV as a function of Cu interlayer thickness for Co(001)/Cu(001)/Al_2O_3/$Ni_{80}Fe_{20}$ junctions. The period of the oscillation observed, 11.4Å, is in agreement with the Fermi surface of Cu. From Yuasa *et al*. [103].

3.7 Coulomb blockade effects

One area of research, which has recently been the subject of much work both experimentally and theoretically, has focused on the interplay between spin-dependent tunneling and the Coulomb blockade in ferromagnetic granular films, double junctions, and single-electron transistors. If a small grain of a ferromagnetic material is inserted in the insulating barrier, tunneling to the grain is strongly influenced by the charging energy. When an electron tunnels into the grain, the electrostatic energy increases by $e^2/2C$, where C is the capacitance of the grain, and therefore, tunneling is blocked unless the barrier presented by the charging energy is overcome by bias voltage or thermal energy. The discreteness of the electron charge manifests itself as characteristic Coulomb staircases in the current-voltage characteristics of the junction. Recent theories [104, 105, 106, 107, 108] have predicted that the combination of the Coulomb blockade and spin-dependent tunneling can lead to both an enhancement and an oscillatory bias dependence of the TMR.

Realizing a system where these effects can be observed is an experimental challenge, however. Granular systems, such as Co clusters in Al_2O_3 are by far the easiest, and an enhancement of the TMR at low temperatures has been observed [109, 110, 111, 112], but the wide distribution of cluster sizes, and hence charging energies, tends to smear out the predicted oscillatory behavior of the TMR. Recently, however, this problem has been addressed by depositing a granular film in a nano-scale constriction, such that the number of clusters within the measured region is small. By additionally gaining better control over the size distribution of the clusters, the predicted oscillatory behavior of the TMR has recently been observed in both the CIP [112] and CPP [113] geometries. In the former case, a clear enhancement of the TMR was observed just above the Coulomb blockade threshold voltage, though the oscillation was heavily damped due to relatively large lateral dimensions. In the latter case, however, the CPP geometry afforded much smaller dimensions, and both an enhanced TMR above the threshold voltage as well as strong oscillatory behavior were observed. While the qualitative behavior of the TMR oscillation with bias voltage is in agreement with theory, most recent theories [104, 105, 106, 108] predict that the TMR remains positive for all biases, while the CPP experiments observe a clear sign change of the TMR. One possible origin for the sign change of the TMR is due to spin accumulation within the clusters, as predicted by Imamura *et al*. [107]. In any case, the significantly enhanced and oscillatory TMR in the Coulomb blockade region is a clear verification of the interesting interplay between the Coulomb blockade and spin-dependent tunneling, which should stimulate further interest in this growing area of research.

4. Models for perfect junctions

A realistic description of SDT requires taking into account accurate atomic, electronic and magnetic structure of MTJs. In general, the quantitative description is rather complicated because transport properties depend exponentially on the properties of the barrier, such as the potential barrier height and the barrier thickness, and are very sensitive to the interfacial roughness, impurity states in the barrier and other types of disorder. In this section we consider perfect tunnel junctions. We ignore, therefore, any type of electron scattering, which can affect tunneling conductance, thereby assuming purely ballistic transport over the whole MTJs. The influence of disorder will be discussed in Sec.5.

We focus on MTJs which are periodic in the plane parallel to the ferromagnet/barrier interfaces. The assumption of the transverse periodicity of a MTJ simplifies significantly the calculation of transport properties. In this case the electron transverse momentum $\mathbf{k}_{\|}$ is conserved, and the tunneling conductance can be represented as the sum over $\mathbf{k}_{\|}$. For the analysis of the conductance it is convenient to use the Landauer-Büttiker formula [114] which, in this case, has the form

$$G = \frac{e^2}{h} \sum_{\mathbf{k}_{\|}} T(\mathbf{k}_{\|}), \qquad (11)$$

where G is the conductance per spin channel, $T(\mathbf{k}_{\|})$ is the transmission coefficient, and the summation is performed over the two-dimensional Brillouin zone. The calculation of the transmission coefficients depends on the particular model which is used for the description of a MTJ. Below we consider, first, simple free-electron models and then analyze more sophisticated approaches which include a multiband electronic structure of the ferromagnets and the barrier.

4.1 Free-electron models

A simplest insight into TMR can be obtained within a free-electron model by assuming a rectangular potential barrier for tunneling. Within this model the exchange splitting of the free-electron bands can be included by considering different potentials for the up- and down-spin electrons, $V_{\uparrow}$ and $V_{\downarrow}$. For electrons tunneling between two identical ferromagnetic electrodes across the rectangular barrier of potential U and thickness d, which is assumed to be not too small, the transmission coefficient per spin is given by (e.g., refs. [19, 115]):

$$T(k_{\parallel}) = 16\kappa^2 \frac{k_1}{\kappa^2 + k_1^2} \frac{k_2}{\kappa^2 + k_2^2} e^{-2\kappa d}, \tag{12}$$

where $k_i = \sqrt{(2m/\hbar^2)(E - V_{\uparrow,\downarrow}) - k_{\parallel}^2}$ is the spin component of the wave vector normal to the interfaces in the ferromagnets (designated by index i = 1, 2) at the Fermi energy E_F, and $\kappa = \sqrt{(2m/\hbar^2)(U - E_F) + k_{\parallel}^2}$ is the decay constant inside the barrier.

In the limit of a thick barrier only electrons which are characterized by the smallest decay constant κ; i.e. those propagating normal to the interface with $k_{\parallel}=0$, contribute to the tunneling current. In this limit the spin polarization of the conductance is given by Slonczewski's formula (10). For not too thick barriers the SP depends on the barrier thickness due to the redistribution of tunneling electrons in the $\mathbf{k}_{\parallel}$ space. This fact was shown in model calculations of MacLaren *et al*. [115], who illustrated the sensitivity of the TMR ratio to the barrier height and thickness. These calculations demonstrate that even within the simplest free-electron description the SP and TMR are determined not only by characteristics of the ferromagnets alone but depend on the properties of the barrier.

Free-electron models were used by a number of researchers to predict magnetoresistive properties of MTJs with paramagnetic layer(s) inserted at the ferromagnet/insulator interface(s) [116, 101, 117]. Vedyayev *et al*. [101] found that the conductance and TMR oscillate with the thickness of the inserted layer. These oscillations are the consequence of quantum-well states in the paramagnetic layer resulting in quantum interference of electron waves. They also demonstrated that a large enhancement in the TMR value could be achieved when two paramagnetic layers at the two interfaces have same thickness. The quantum oscillations of TMR with the thickness of a non-magnetic layer were also found by Mathon and Umerski [102], who used realistic tight-binding bands of Co and Cu to calculate the TMR in the presence of a Cu layer inserted at the Co/vacuum interface. Possible enhancements of TMR due to quantum-well states were predicted within free-electron models for double-barrier junctions in which a third ferromagnetic layer is inserted within the barrier [118, 119, 120].

In order to observe these quantum effects electrons should preserve their coherence in the tunneling process. Intermixing between transport modes with different transverse momenta $\mathbf{k}_{\parallel}$ due to scattering by disorder and impurities could destroy the predicted behavior. Zhang and Levy [116] proposed that the rapid drop of TMR observed for most MTJs with inserted non-magnetic layers at the interfaces is due to the loss of coherence in transmission through these layers which is caused by fluctuations in the inserted layer thickness. The recent observation of quantum oscillations of TMR in Co/Cu/Al_2O_3/$Ni_{80}Fe_{20}$ tunnel junctions with epitaxial Co/Cu(001) layers by Yuasa *et al*. [103] (see also Sec.3.6) indicates that the quantum coherence can be preserved in real MTJs.

Although free-electron models capture some important features of spin-dependent tunneling, they cannot be used for the quantitative description of TMR. In particular, results of the free-electron consideration are very sensitive to the profile of the potential barrier [121]. Moreover, free-electron models ignore the multiband electronic structure of the ferromagnetic electrodes and the ferromagnet/insulator interfaces. Finally, the free-electron models do not take into account the complex band structure of the insulator that, as we will see in the Sec. 4.3, is decisive for selecting bands which tunnel most effectively across the barrier. All these arguments demonstrate that only using accurate description of the electronic structure of the entire MTJ it is possible to make quantitative conclusions about TMR.

4.2 Bonding at the ferromagnet/insulator interface

One of the important properties of MTJs, which affects strongly the spin-dependent tunneling, is the chemical bonding at the ferromagnet/insulator interface. The bonding mechanism determines the effectiveness of transmission across the interface which can be different for electrons of different character. Tsymbal and Pettifor [75] showed that for tunneling from transition-metal ferromagnets across a thin barrier layer the spin polarization of the conductance depends strongly on the interfacial bonding between the ferromagnet and the insulator. They found that in case of the $ss\sigma$ bonding the spin polarization of the conductance is positive which is in agreement with experimental data on tunneling through an alumina spacer [6]. Increasing the $sd\sigma$ bonding at the interface, however, reduces the spin polarization and can even lead to a change in its sign.

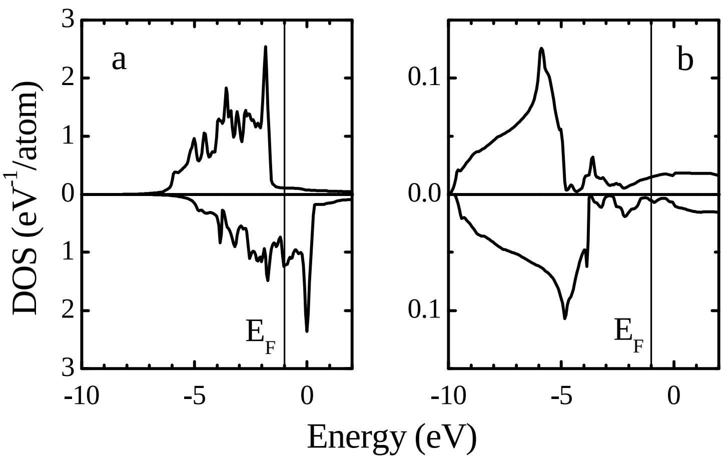

Fig.13 Density of states for bulk fcc Co projected to the d orbitals (a) and the s orbitals (b) for the majority-spin electrons (top panels) and minority-spin electrons (bottom panels). The spin polarization of the d-DOS at the Fermi energy is opposite to that for the s-DOS. Note a different scale in (a) and (b).

This conclusion can be explained by the fact that in the presence of the interfacial ssσ bonding only *s* states of the ferromagnet are coupled with those of the insulator. In this case only *s* electrons of the ferromagnetic layer can contribute to the tunneling current. It is known, however, that the *s* component of the density of states (DOS) is suppressed within the d band of the 3d metals due to the strong *s-d* hybridization. This is demonstrated in Fig.13a,b which shows the DOS projected to the d- and s-orbitals for bulk fcc Co. As is evident from the figure, although the d-DOS at the Fermi energy is lower for the majority spins than that for the minority spins, the s-DOS is higher making the spin polarization positive. Increasing the sdσ bonding at the interface, however, results in a large contribution of the d electrons to the tunneling current. In this case, due to the interfacial $sd\sigma$ bonding, the d states of the ferromagnet can evolve into the s states of the insulator and can be transmitted across the MTJ. The negative spin polarization of the d-DOS at the Fermi energy (see Fig.13a) can then be reflected in the tunneling current.

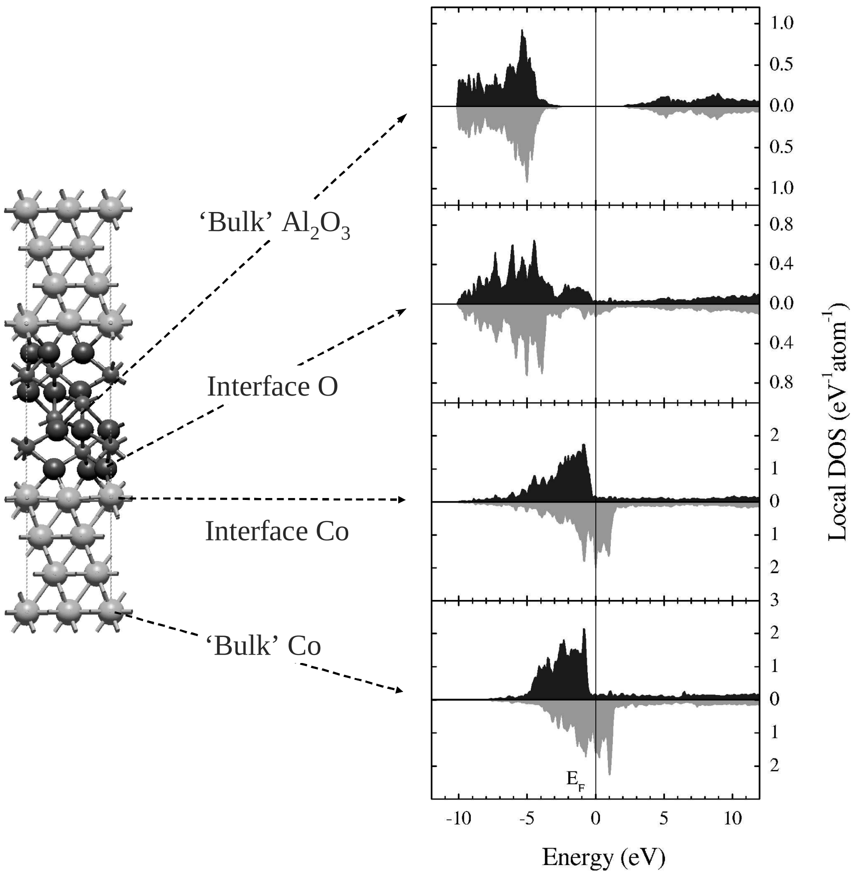

Fig.14 The calculated atomic structure and local density of states (DOS) for majority-spin electrons (top panels) and minority-spin electrons (bottom panels) for a $Co/Al_2O_3/Co$ tunnel junction. After Oleinik *et al*. [127].

The effect of bonding at the ferromagnet/insulator interface was proposed to explain the experimentally observed inversion of the spin polarization of tunneling electrons from Co across a $SrTiO_3$ barrier [9] (see Sec.3.4). The bonding mechanism was also put forward to explain positive and negative values of TMR depending on the applied voltage in MTJs with Ta_2O_5 and Ta_2O_5/Al_2O_3 barriers [41] and to elucidate the inversion of TMR observed in Co-contacted multiwalled carbon nanotubes [122]. Itoh and Inoue predicted theoretically the strong sensitivity of the magnitude of TMR to the sp-d mixing at the ferromagnet/alumina interface in the presence of imperfectly oxidized Al or O ions [123]. Tsymbal *et al*. [124] found that oxygen deposited on the surface of Fe inverts the spin polarization of the density of states at the Fermi energy propagating in vacuum, due to hybridization of the iron 3*d* orbitals with the oxygen 2*p* orbitals and the strong exchange splitting of the antibonding oxygen states. Earlier *ab-initio* calculations of the electronic structure of a Co/HfO_2 tunnel junction [125] demonstrated the inversion of the spin polarization at the Fermi energy. For $Co/SrTiO_3/Co$ tunnel junctions, Oleinik *et al*. [126]

predicted that the exchange coupling between the interface Co and Ti atoms mediated by oxygen induces a magnetic moment of 0.2 μ_B on the interface Ti atom, which is aligned antiparallel to the magnetic moment of the Co layer. All these findings indicate an important role of the bonding at the ferromagnet/insulator interface in spin-dependent tunneling.

However, despite the importance of the interfacial bonding, this bonding alone is *not* able to explain the positive spin polarizations of electrons tunneling from 3d ferromagnets across an alumina barrier observed experimentally. First-principle calculations of the electronic structure of a Co/Al_2O_3/Co tunnel junction [127] demonstrate the presence of a strong covalent bonding between d-orbitals of Co and p-orbitals of O at the Co/Al_2O_3 interface. This can be seen from Fig.14 which shows the local densities of states corresponding to different atoms in the Co/Al_2O_3/Co tunnel junction. As is evident from the local DOS for the interfacial O atom, the hybridization of the Co-3d states and O-2p states leads to formation of the bonding and antibonding states. The oxygen antibonding states are exchange-split mirroring the strong exchange splitting of the Co-3d states. Although the calculations find a negative spin polarization in the local DOS at the Fermi energy on the O and Al atoms close to the interface (which reflect the negative spin polarization of the DOS in Co), the spin polarization of the DOS becomes positive on interior atoms of the alumina layer [127]. This indicates that the spin polarization of the tunneling current should also be positive which is in agreement with the experiments on spin-dependent tunneling. In order to explain this behavior one needs to consider explicitly the mechanism of conductance in MTJs and identify those bands dominating the tunneling process. This will be the subject of our discussion in Sec. 4.3.

4.3 First-principle calculations of TMR

First-principle methods based on density functional theory within the local spin density approximation (LSDA) for the electronic structure and the Landauer-Büttiker formula (11) for the conductance provide the basis for an accurate calculation of spin-dependent tunneling in MTJs. This approach is advantageous due to a realistic, multiband description of the electronic structure, which takes into account the character and the spin polarization of the electronic states in ferromagnetic electrodes, the interfacial localized states, the variation of the potential across the barrier, and the evanescent states in the insulator.

Using the layered Korringa-Kohn-Rostoker (LKKR) method MacLaren *et al*. [128] calculated the electronic structure and the tunneling conductance in Fe/ZnSe/Fe(100) MTJs. They found that the spin asymmetry in the conductance increases dramatically with the increasing barrier thickness. They showed that the difference in the decay rates for the majority and minority spin channels follows from the symmetry of the Bloch states at the Fermi energy, which have different spin injection (extraction) efficiency and different decay rates when tunneling across the barrier. Since at a relatively large barrier thickness the conductance is dominated by states at $\mathbf{k}_{\|} = 0$, they analyzed the results in terms of the layer resolved density of states (DOS) within the electrodes and the tunneling barrier at $\mathbf{k}_{\|} = 0$, as shown in Figs.15a,b. As is evident from these figures, there are three decay rates, which are associated with the angular momentum character of the bands within the barrier. The rate of the decay is slowest for bands with *s* character and most rapid for bands with only *d* character. In addition to the different decay rates, the ease of injection and extraction depends upon the character of the band in the electrode. For example, in the majority channel, the Δ_1 band, because it is compatible with the *s* character, couples efficiently with a decaying *sp* state in the ZnSe, and thus, this band dominates the conductance. The much smaller tunneling conductance seen for the minority spins in Fig.15b is a direct result of there being no Δ_1 band present at the Fermi energy.

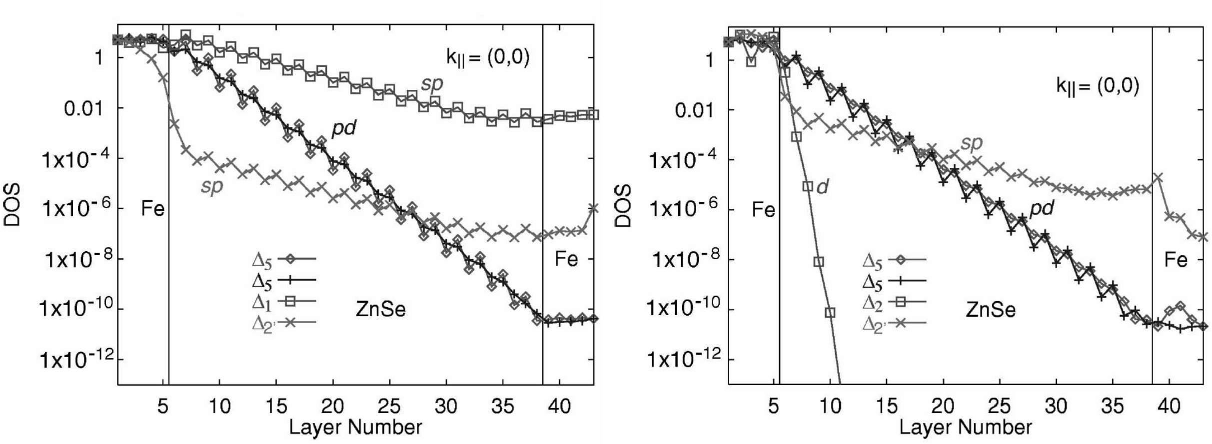

Fig.15 The calculated layer-dependent density of states (DOS) for the majority (a) and minority (b) Bloch states at $\mathbf{k}_{||}=0$ for Fe/ZnSe/Fe junction. Different decay rates and injection efficiencies for the states of different character and symmetry are seen. After MacLaren *et al*. [128].

Based upon these results, MacLaren *et al*. [128] concluded that the expected spin dependence of the tunneling current can be deduced from the symmetry of the Bloch states at the Fermi energy. The bands with *s* character are able to couple most efficiently across the interface, and decay most slowly in the barrier. For Fe, Co, and Ni ferromagnets the majority states at the Fermi energy have more *s* character than the minority states, which tend to have mainly *d* character. Thus, the majority conductance is expected to be greater than the minority conductance resulting in a slower decay with the barrier thickness for the former. These conclusions are expected to be also valid for MTJs with an Al_2O_3 barrier which is consistent with the experimental observations [6]. This explains an earlier hypotesis of Stearns [7] who proposed that most dispersive bands are desisive for the tunneling process.

MacLaren *et al*. [128] pointed out that the spin asymmetry in the tunneling conductance should depend on the substrate crystal face. In the case of Fe, for example, an examination of the band structure shows that for [100], [111], and [110] directions all have majority bands with *s* character present, and for all but the [100] direction, a band with this symmetry also crosses the Fermi energy for the minority channel. Thus, the [100] direction should exhibit the largest asymmetry in tunnel conductance. Indeed, the dependence of TMR on the crystal face of the epitaxial Fe electrodes in Fe/Al_2O_3/CoFe junctions was recently observed by Yuasa *et al*. [67] (see Sec.3.4). However, they found that larger values of TMR for tunneling from Fe(110), rather than from Fe(100) electrodes. Still, further analysis of bands contributing to tunneling is needed to obtain consistency between the theory and experiment.

Mavropoulos *et al*. [129] emphasized the importance of the evanescent gap states in the tunneling barrier for spin-dependent tunneling. They used a notion of the complex band structure to analyze the metal-induced gap states with specific examples of Si, Ge, GaAs, and ZnSe. Using the empirical pseudopotential method Mavropoulos *et al*. calculated the complex band structure of these semiconductors, which enabled them to determine the decay rate parameter $\kappa(\mathbf{k}_{\|},E)$ and the symmetry of the evanescent states. They found that the states which belong to the identity representation Δ_1 should have minimum decay rates for a broad class of materials. For semiconductors (insulators) with a direct band gap (such as GaAs, ZnSe or semiconductors with a higher atomic number or/and ionicity) these states are centered on the Γ point ($\mathbf{k}_{\|} = 0$). However, for indirect band gap semiconductors (such as Si and Ge) these states might be centered on other points in the Brillouin zone depending on the position of the Fermi level with respect to the bottom of the conduction band. Thus, it was demonstrated that the complex band structure of the barrier material allows explaining very important features of the tunneling process and can be regarded as one of the fundamental characteristics of spin-dependent conductance in MTJs.

The conclusions drawn from considering the complex band structure are also applicable to oxide barrier MgO [129]. Recently Butler *et al*. [82] calculated the electronic structure and the tunneling conductance in Fe/MgO/Fe MTJs within the approach of their previous work [128]. Their conclusions essentially support findings reported in refs. [128] and [129]. In particular, they found that due to the absence of the minority Δ_1 band at the Fermi energy of Fe (100), the majority-spin conductance dominates tunneling which leads to a very high TMR for thick enough barriers. Mathon and Umerski [130] arrived at the same qualitative conclusions, after calculating the TMR in Fe/MgO/Fe MTJs using the multiband tight-binding description for the electronic structure. Although not as accurate as first-principle based theory, this approach is far more realistic than free-electron models, as demonstrated in the modeling of GMR [18]. Earlier, the tight-binding method was used by Mathon [131], who attempted to unify the description for the CPP GMR and TMR phenomena.

In conclusion of this section, we note that the experiments performed on epitaxial Fe/MgO/FeCo MTJs [81] (see also Sec.3.5) show much smaller values of TMR compared to those predicted theoretically. This might be due to the formation of a partially oxidized FeO layer at the interface which was found in the experiments by Meyerheim *et al*. [132]. In addition, effects of disorder which are ignored in the theoretical studies of refs. [82, 131] may play a significant role. Epitaxial Fe/ZnSe/FeCo magnetic tunnel junctions show less impressive behavior demonstrating a sizable magnetoresistance of 16% only at low temperatures [133]. This is obviously the consequence of a semiconducting nature of the ZnSe barrier which makes the mechanism of conductance to be different from that considered theoretically. The presence of impurity/defect states in the electrodes and in the barrier makes the ballistic approach inadequate for the description of spin-dependent tunneling in these junctions. The effects of disorder will be discussed in the next section.

5. Models for disordered junctions

Actual tunnel junctions contain large amounts of disorder in the electrodes, in the barrier and at the electrode/barrier interfaces. This disorder may represent interdiffusion at the interfaces, interface roughness, impurities, and defects such as grain boundaries, stacking faults, and vacancies. Interdiffusion dramatically changes the electronic and atomic structure, which affects TMR in a critical way (e.g., [134, 135, 136]). Interface roughness leads to fluctuations in the barrier thickness that strongly alters tunneling conductance [137]. Impurities and defects in the barrier introduce complex mechanisms that assist tunneling [38, 44, 138, 139, 140, 141, 142].

This is especially important in the case of amorphous barriers, although even in epitaxially-grown tunnel junctions the effects of disorder might be decisive. Disorder in the electrodes mixes bulk and interface states and thereby influences TMR [143, 144, 145]. In this section we consider some consequences of disorder in MTJs that are important for the understanding of experiments on spin-dependent tunneling.

5.1 Contribution of interface states

As we saw, for perfect tunnel junctions it is important to identify the propagating (bulk) states in the ferromagnets which are coupled to the slowest decaying state in the barrier, and therefore dominate tunneling. However, as was pointed out by Levy *et al*. [143], calculations made for purely ballistic transport over the whole junction cannot be directly compared to data on real junctions, because the ballistic conductance underestimates a contribution from states localized near the interfaces. In disordered tunnel junctions these interface states are coupled to the propagating states in the electrodes by diffusive and relaxation processes, which provide additional conduction channels. Under these conditions the electronic structure at the electrode/barrier interfaces may control the tunneling current.

Although, the interface states can contribute to the tunneling conductance even in perfect magnetic tunnel junctions, their contribution is normally small. The interface states from both sides of the barrier are coupled to the propagating states from the other side of the barrier and are coupled to each other. This leads to a resonant mechanism of tunneling which manifests itself as spikes in the conductance distribution at particular $\mathbf{k}_{||}$ points in the two-dimensional Brillouin zone [146]. The width of these spikes is determined by the strength of the coupling through the barrier, which decreases exponentially with the barrier thickness.

The presence of defect scattering in the electrodes and at the electrode/barrier interfaces makes the coupling between the interface and bulk states much more efficient. This affects the spin polarization of tunneling electrons due to different spin polarizations of the bulk and interface states. Fig.16 shows a model tight-binding calculation of the conductance across a disordered Fe/I/Fe tunnel junction, in which the insulator (I) is described by a simple tight-binding band that provides no states at the Fermi energy. Disorder is introduced by random variations in the on-site atomic energies of width γ within 10 monolayers of the Fe electrode adjacent to the interface. As is evident from the figure, in the absence of disorder ($\gamma = 0$) the spin polarization is large, about 0.9. This high value of the spin polarization is consistent with calculations of Butler *et al*. [82] and Mathon and Umerski [130] for Fe/MgO/Fe MTJs and reflects the presence of the Δ_1-symmetry band at the Fermi energy for the majority-spin electrons of Fe(100) and the absence of such a band for the minority-spin electrons. With increasing disorder the conductance of the minority spins increases dramatically, whereas the conductance of the majority spins is insensitive to disorder, which leads to a decrease in the spin polarization. The enhancement in the transmission for the minority-spin electrons with disorder is due to the interface states which have a strong weight at the Fermi energy in Fe (see, e.g., ref.[144]). These interface states get coupled to the bulk states within the same electrode and thereby contribute to the conductance. We note that, in addition, disorder breaks the symmetry of the system and mixes the propagating Bloch states in the leads. This makes it possible for the states which are not able to tunnel effectively through the barrier in the perfect tunnel junction by symmetry to be mixed with the states which are and therefore to be involved in transport.

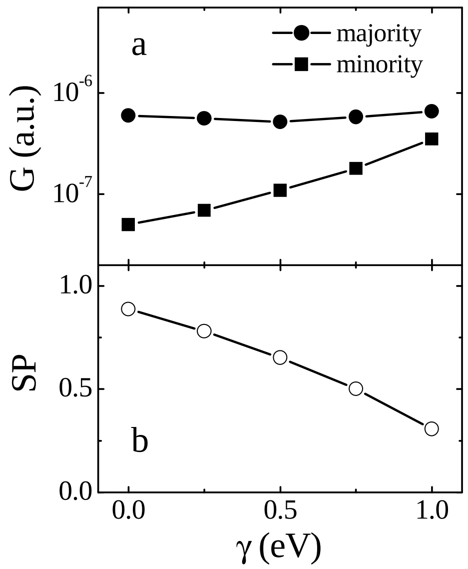

Fig.16 The calculated conductance for the majority- and minority-spin electrons (a) and the spin polarization (b) of the conductance in a Fe/I/Fe tunnel junction as a function of disorder parameter γ. With increasing disorder the SP decreases due to the contribution from the interface state in Fe. A typical value of γ for sputtered 3d-metal films is about 0.5eV. After Tsymbal [147].

We see, therefore, that disorder makes the electronic structure at the electrode/barrier interfaces to control the tunneling current. Disorder leads to diffuse scattering that couples the propagating states in the bulk of the electrodes to the states localized at the interfaces. This mechanism is the fundamental origin of the decisive role of the interface electronic structure, which was demonstrated experimentally by LeClair *et al*. [10, 11, 12] (see Sec.3.6).

5.2 Effect of disorder in the barrier

Disorder in the barrier layer has a dramatic effect on spin-dependent tunneling. The presence of disorder broadens the conduction and the valence bands of the insulator and creates localized electronic states within the band gap. The broadening of the bands reduces the effective potential barrier for tunneling that, according to Slonczewski [19] (Sec.2.4), influences negatively the spin polarization of tunneling electrons in MTJs.

Even more decisive effects occur due to the formation of localized impurity/defect states in the barrier. If the energy of these states is close to the Fermi energy, they lead to resonance tunneling. In order to understand the consequences of resonant tunneling in MTJs we consider a simple one-dimensional model for impurity-assisted tunneling. In this case the conductance per spin is given by

$$G=\frac{4e^2}{h}\frac{\Gamma_1\Gamma_2}{\left(E-E_r\right)^2+\left(\Gamma_1+\Gamma_2\right)^2}, \tag{13}$$

where E_F is the Fermi energy, E_r is the energy of the resonant state and Γ_1 and Γ_2 are leak rates of an electron from the impurity state to the left and right electrodes. We assume for simplicity that the latter are proportional to the densities of states of the electrodes, ρ_1 and ρ_2, at the left and right interfaces, so that $\Gamma_1 \propto \rho_1 \exp[-2\kappa x]$ and $\Gamma_2 \propto \rho_2 \exp[-2\kappa(d-x)]$, where κ is the decay constant and x is the position of the impurity within the barrier of thickness d [148]. Off resonance, when $\left|E-E_r\right| >> \Gamma_1+\Gamma_2$, the latter assumption implies that the spin conductance is given by $G \propto \rho_1\rho_2$. When tunneling occurs between ferromagnetic electrodes this leads to TMR, which is given by Julliere's formula (7), with P_i (i = 1, 2) given by Eq.(8).

In order to take into account disorder in real tunnel junctions, the conductance should be averaged over the energy and position of impurities. Following ref.[138], we assume for simplicity a homogeneous distribution of impurities with uniform density $D(E_r)=\delta$. Integrating Eq.(13) with respect to the impurity position and energy we obtain for non-halfmetallic electrodes ($\rho_i^{\uparrow\downarrow} \neq 0$) and for not too thin barriers ($\exp[-\kappa d] \ll 1$) that

$$\left\langle G\right\rangle \simeq \frac{e^2}{h}\frac{\delta e^{-\kappa d}}{2\kappa d}\sqrt{\rho_1\rho_2}\ . \tag{14}$$

For ferromagnetic electrodes this implies that the respective spin polarization of the tunneling conductance across a disordered barrier is reduced compared to that for a perfect barrier so that

$$.\ P_i=\frac{\sqrt{\rho_i^{\uparrow}}-\sqrt{\rho_i^{\downarrow}}}{\sqrt{\rho_i^{\uparrow}}+\sqrt{\rho_i^{\downarrow}}}, \tag{15}$$

where i = 1, 2. This leads to a diminished value of TMR, which is still given by Julliere's formula (7), but the effective spin polarizations of the electrodes are defined by Eq.(15). For example for $\rho^{\uparrow\downarrow}=\rho_1^{\uparrow\downarrow}=\rho_2^{\uparrow\downarrow}$ and $\rho^{\uparrow}/\rho^{\downarrow}=3$, we obtain P=50% and TMR=67% for a perfect junction, whereas we obtain P=27% and TMR=15% for a disordered junction. This significant reduction in the SP and TMR is the consequence of spin-dependent leak rates, Γ_1 and Γ_2, and the inversion of magnetoresistance at resonant conditions, as we will see in Sec.5.3.

In real MTJs with amorphous barriers the situation is more complicated because of the contribution from multiple resonances resulting from the interference of electrons scattered by several localized stated in the barrier. Tsymbal and Pettifor [140] found that in strongly disordered tunnel junctions the tunneling current flows through a few regions of the insulator where local disorder configuration provides highly conducting channels for electron transport. This mechanism of conduction leads to a broad distribution of the tunneling current, which is in agreement with experimental data on local transport properties of Al_2O_3 tunnel barriers [137]. Tsymbal and Pettifor predicted a decrease in the spin polarization of the tunneling current with disorder and insulator thickness. Interestingly, they found that the TMR is in agreement with the Julliere's formula (7), in which $P_{1,2}$ is defined as the spin polarization of the tunneling current from the ferromagnet to a non-magnetic metal. This might explain the success of the Julliere's formula when comparing the TMR magnitudes with the SP values measured in experiments on superconductors (Sec.3.4).

Several authors addressed the problem of the influence of magnetic impurities within the barrier on TMR [44, 141, 142]. In particular, Vedyayev *et al*. [44] found that at low temperatures and zero bias voltage, the TMR in a MTJ with paramagnetic impurities can be larger than that of the same structure without paramagnetic impurities. They also found that an increase in temperature leads to a decrease in the TMR magnitude due to the excitation of spin-flip processes resulting in mixing of spin-up and down channels. Jansen and Lodder [141] showed that for spin-polarized states in the barrier, the magnetoresistance due to resonant tunneling can be enhanced compared to the magnetoresistance due to direct tunneling. Inoue *et al*. [142] extended the treatment of the tunnel conductance to take into account many-body effects of the exchange interaction between the tunneling electrons and localized spins. They found that the TMR ratio decreases by approximately 10% due to the spin-flip process caused by the exchange interaction.

5.3 TMR at resonant conditions

Eq. (13) for resonant tunneling predicts a strong variation of TMR as a function of the impurity energy near the resonance (similar to that shown in Fig.17c). Exactly at the resonance, i.e. when $E_F - E_i = 0$, the magnetoresistance is inverted. Indeed, assuming for simplicity an asymmetric position of impurity we obtain from Eq.(13) that $G \propto \rho_2 / \rho_1$, if $x < d/2$ and hence $\Gamma_1 \gg \Gamma_2$, and we obtain that $G \propto \rho_1 / \rho_2$, if $x > d/2$ and hence $\Gamma_1 \ll \Gamma_2$. In both cases, the conductance is inversely proportional to the density of states of the one of the ferromagnets which results in the sign inversion of the TMR:

$$TMR = -\frac{2P_1P_2}{1+P_1P_2} \tag{16}$$

(compare to Eq.(7)). As we see, the inversion of TMR originates from the spin-dependent leak rates that under resonant conditions invert the effective SP of the one of the ferromagnetic electrodes.

The question arises, whether it is possible to observe the strong variation and the inversion of magnetoresistance at resonant conditions. As we saw in the previous section, the averaging over a large number of disorder configurations corresponding to different energies and positions of impurities simply results in the suppression of TMR. This is due to a relatively large area of thin-film tunnel junctions, which normally spans values from a fraction of μm^2 to a few mm^2. Very recently Tsymbal *et al*. [149] found that it is possible to reveal effects dominated by a single localized state and to observe a broad distribution of TMR values including the inverse magnetoresistance in magnetic nanojunctions with a small cross section. As was demonstrated earlier by Doudin *et al*. [150], nanowire junctions grown by electrodeposition with a cross section less than $0.01 \mu m^2$ display two-level fluctuations of the electric current which indicate an impurity/defect-driven transport. By performing measurements on a large number of samples and comparing experimental and calculated statistics Tsymbal *et al*. [149] showed that the TMR is inverted when the energy of a localized state in the barrier matches the Fermi energy of the ferromagnetic electrodes. The experimental and calculated distributions of TMR along with the predicted energy dependence of magnetoresistance for a tunnel junction demonstrating the inverse TMR are displayed in Fig.17.

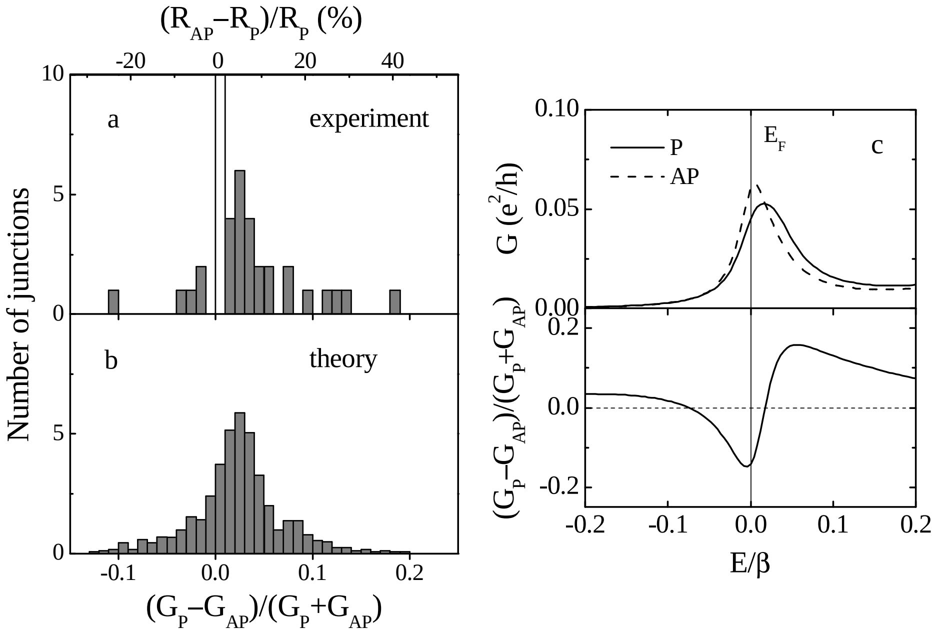

Fig.17 Experimental (a) and calculated (b) distribution of magnetoresistance in magnetic Ni/NiO/Co nanojunctions and (c) predicted conductance for parallel (P) and antiparallel (AP) configuration of the electrodes (top) and TMR as a function of energy for a tunnel junction which shows the inverse TMR (bottom). The highest positive and negative values of magnetoresistance measured are +40% and −25%. The unshaded bar indicates a possible contribution from samples with multiple junctions. After Tsymbal *et al.* [149].

Another possible way to observe the predicted strong variation of TMR due to resonant tunneling is to use local characterization techniques such as STM [79] and BEEM [151]. As was shown by Tsymbal and Pettifor [152], in this case a local impurity/defect state in the barrier can be detected due to electrons traversing ballistically the top metallic layer and, then, tunneling resonantly across the barrier via the localized electronic state in the band gap of the insulator. They found that the TMR magnitude varies dramatically as the tip scans the area above the impurity atom. If the tip is located directly above the impurity the TMR is inverted. This phenomenon could be observed by the STM or BEEM techniques provided that the switching of the magnetic alignment of the two electrodes in a MTJ is achieved. Note that the BEEM technique [151] has the advantage of the intrinsic ballistic nature of the transited current, whereas the STM technique [79] requires the use of high-quality epitaxial junctions.

6. Conclusions

Stimulated by the discovery of giant magnetoresistance (GMR) in metallic magnetic multilayers (for a recent review on GMR see ref.[18]) spin electronics has developed into a vigorous field of research. Spin-dependent tunneling in MTJs, one of the areas of spin electronics, has aroused considerable interest due to a large room temperature magnetoresistance. Significant progress in the fabrication and characterization of MTJs and in the understanding of basic mechanisms which control the spin polarization of tunneling electrons have been achieved in the past few years. This has led to important advances towards the applications of magnetoresistive devices based on MTJs, such as random-access memories (e.g., refs.[153, 27]), as well as elucidating the fundamental physics that governs the functioning of these devices.

One of the breakthroughs in the understanding of TMR is the recognition of the fact that the tunneling spin polarization in MTJs is determined not only by the properties of the ferromagnets alone but depends on the atomic and electronic structure of the entire junction including the insulator and the ferromagnet/insulator interfaces. This fact broadens dramatically the possibilities for altering the properties of MTJs. In particular, by modifying the electronic properties of the tunneling barrier and the ferromagnet/insulator interfaces it is possible to engineer MTJs with properties desirable for device applications.

There has been a significant progress in the theoretical description of TMR. First-principle calculations of the electronic structure and the conductance in MTJs have led to deeper insights into the role of symmetry of the electronic states of the ferromagnets and their coupling to the evanescent states in the barrier. These calculations have also demonstrated the importance of the interfacial states and the character of chemical bonding across the metal/insulator interfaces. More can be expected from ab-initio models in addressing the role of defects at the interfaces such as partially oxidized Fe layer in Fe/MgO/Fe epitaxial junctions [132]. In general, including defect scattering is an important ingredient for further progress in the theoretical description of TMR. However, a proper first-principle treatment of all the existing defects might be too expensive computationally and, therefore, reliable simplified models become of great importance.

The voltage dependence of TMR is far from being completely understood. The difficulty arises from the fact that a number of different processes may contribute to the voltage dependence, such as the spin-dependent electronic structure of the electrodes, inelastic scattering by defect/impurity states in the barrier, and electron-phonon, electron-magnon and electron-electron interactions. For a realistic theoretical description of the voltage dependence, it might be necessary to incorporate the non-equilibrium Green's function formalism into the theory of TMR [154]. Further studies have to be performed to elucidate the role of different mechanisms in the voltage dependence.

There is unrealized potential of half-metallic-based tunnel junctions. Ideally incorporation of 100% spin-polarized ferromagnets into a MTJ should lead to an infinitely large TMR. Unfortunately, experiments so far show an unimpressive behavior of MTJs based on half metals at room temperature. A possible reason for this is the poor quality of the half-metal/barrier interfaces, resulting in a dramatic reduction of the spin polarization and/or the detrimental effect of thermal fluctuations on the spin polarization [155]. More work has to be done to obtain well-controlled half-metallic films and interfaces with the robust spin polarization.

The success of spin-dependent tunneling in MTJs has exposed a number of new directions for a further research. In particular, spin-electronics applications based on magnetotransport phenomena in semiconductors have recently started to attract more and more attention. Making use of semiconductors in spin electronics has the advantage of incorporating the magnetoresistive devices into existing semiconductor technologies. The feasibility of using semiconductors is supported by their capability to carry highly spin-polarized currents over long distances [156] and by the successful demonstration of electrical spin injection from magnetic dilute semiconductors [157, 158]. Recent discoveries of room-temperature spin injection from metallic Fe into GaAs [159, 160], a large magnetoresistance in GaMnAs/AlAs tunnel junctions [161], and the possibility of spin filtering across ferromagnet/semiconductor interfaces at room temperature [162] stimulated further interest in this field. The advances in the understanding of mechanisms controlling quantum magnetotransport in MTJs are expected to be very instrumental in achieving progress in this very rapidly developing area of spin electronics.

Another example is the ballistic conductance in magnetic nanocontacts. Experiments performed on nanocontacts fabricated from Ni nanowires have shown sizable values of magnetoresistance at room temperature [163]. These experiments have been explained by the domain wall constrained by the nanocontact region [164, 165]. Very recent studies on electrodeposited Ni nanocontacts found, however, that it is possible to achieve magnetoresistance as high as 3000% at room temperature [166], which can hardly be explained only by spin-dependent scattering by the constrained domain wall. The physical mechanism causing this phenomenon is, at present, unknown and further studies, both theoretical and experimental, are desirable.

Elucidating spin-dependent conductance in metallic nanocontacts is also important for the understanding of the electronic transport through pinholes in magnetic tunnel junctions. As was discovered recently [167], in some cases pinholes in a tunnel barrier may mimic tunneling and make it difficult to distinguish between electron conduction through pinholes and direct tunneling. Recent experiments showed that it is possible to observe 15% magnetoresistance at room temperature from electrodeposited nanocontacts through pinholes in MTJs [168].

Using spin-polarized barriers in MTJs may be promising for applications as spin filters. The demonstration of highly efficient spin-filter tunneling using EuS [94] should stimulate further research in this field, in particular, towards the search for spin-polarized barriers which preserve their properties at room temperature. Next-generation devices based on spin-filter tunnel structures have potential for highly efficient spin injection into semiconductors [169], essential for the development of semiconductor-based spin electronics.

In summary, spin-dependent tunneling in magnetic tunnel junctions is a fast-growing area of research, which combines both tremendous technological potential and deep fundamental physics. It has stimulated new directions in spin-dependent electronic transport, which promise exciting results in the future.

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