Bias dependence of tunneling magnetoresistance in magnetic tunnel junctions with asymmetric barriers

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Abstract
The transport properties of magnetic tunnel junctions (MTJs) are very sensitive to interface modifications. In this work we investigate both experimentally and theoretically the effect of asymmetric barrier modifications on the bias dependence of tunneling magnetoresistance (TMR) in single crystal Fe/MgO-based MTJs with (i) one crystalline and one rough interface, and (ii) with a monolayer of O deposited at the crystalline interface. In both cases we observe an asymmetric bias dependence of TMR and a reversal of its sign at large bias. We propose a general model to explain the bias dependence in these and similar systems reported earlier. The model predicts the existence of two distinct TMR regimes: (i) a tunneling regime when the interface is modified with layers of a different insulator, and (ii) a resonant regime when thin metallic layers are inserted at the interface. In the tunneling regime, negative TMR is due to the high voltage which overcomes the exchange splitting in the electrodes, while the asymmetric bias dependence of TMR is due to the interface transmission probabilities. In the resonant regime, inversion of TMR could happen at zero voltage depending on the alignment of the resonance levels with the Fermi surfaces of the electrodes. Moreover, the model predicts a regime in which TMR has different signs at positive and negative bias, suggesting possibilities of combining memory with logic functions.

(Some figures may appear in colour only in the online journal)

1. Introduction
The field of spintronics has been very successful in producing magnetoresistive devices for magnetic memory and sensor applications [1]. Magnetic tunnel junctions (MTJs) came to the forefront of spintronics research after theoretical predictions of very high positive tunneling magnetoresistance (TMR) in Fe/MgO/Fe MTJs [2]. Shortly after, TMR in excess of 200% was reported experimentally in these junctions [3, 4]. More recently TMR as high as 604% at room temperature was reported in MgO-based MTJs with CoFeB electrodes [5]. It has been recognized that the interfaces are crucial for...
the TMR amplitude and voltage dependence, and consequently interface engineering has received a great deal of attention [6, 7].

There are a number of experimental studies of the influence of modified interfaces on the sign and bias dependence of TMR, in particular with insulating layers such as Ta$_2$O$_5$ [8] and NiO [9], metallic layers Cr [10] and Fe$_3$O$_4$ [11], adatoms C [12, 13] and O [14, 15], and morphologically different interfaces [16–19]. A recent experimental work shows that the insertion of a thin NiO layer at one of the interfaces of a CoFe/MgO/CoFe(001) MTJ gives rise to an asymmetric bias dependence of the TMR [9]. Also, switching from positive to negative TMR was observed at larger bias. It was suggested that the effect is due to formation of a non-collinear magnetic structure at the CoFe/NiO interface, however, it is not clear how that fact may affect the bias dependence of TMR. Very similar observations were reported earlier in NiFe/Ta$_2$O$_5$/Al$_2$O$_3$/NiFe MTJs [8]. The sign reversal was interpreted, using the Julli`ere model, in terms of the change of the spin polarization of the electrodes with the bias [20]. Strongly asymmetric TMR bias dependence was also reported. Moreover, essentially identical behavior and sign reversal of TMR was observed in experiments doping the interface with non-magnetic metal layers (Cr) [10], non-magnetic atoms (C) [12, 13], or by just varying the morphology of the interface [16].

First principles transport calculations with finite bias are fairly difficult and therefore not commonplace. There are several density functional theory based calculations of Fe/MgO-based MTJs with ideal and oxidized interfaces at finite bias [18, 21–24]. Some of them consider MTJs with asymmetrically modified interfaces and report asymmetric behavior of TMR [18, 20, 23]. Overall, the asymmetric bias dependence and sign change of TMR emerge as general features of many diverse systems, however, interpretation is usually limited to qualitative arguments based on the Julli`ere model on a case-by-case basis. Further analysis is needed to understand the underlying physics of these phenomena.

In this paper, we report measurements of the bias dependence of TMR in pairs of single crystal Fe/MgO MTJs—one with clean interfaces (Fe/MgO/Fe) and another with a monolayer of O deposited at one of the interfaces (Fe/O/MgO/Fe). Due to the growth procedure, in both cases the bottom (first) interface is atomically sharp while the top (second) is of lower quality [25]. The experiments show that TMR is asymmetric with respect to the voltage sign and negative TMR arises at high voltage. The insertion of an O layer at the higher quality interface acts as an additional barrier and slightly reduces the asymmetry of the bias dependence of TMR [14, 25]. We propose a model which explains these experimental observations as well as other previously reported experiments. The model relates the interface asymmetry to the interface transmission functions. We show that interface modifications which preserve the tunneling regime lead to asymmetric bias dependence and TMR inversion at large bias, while modifications with metallic layers can lead to TMR inversion even at zero bias due to resonant transmission.

2. Experimental results

The details of the experimental procedure have already been discussed in previous papers [14, 17]. The samples were grown on MgO(001) substrate and the MgO barrier thickness was between 1.1 and 2.3 nm (5–11 monolayers). Pairs of samples, Fe/MgO/Fe (A) and Fe/O/MgO/Fe (B), were grown simultaneously by molecular beam epitaxy in the same ultra-high vacuum (UHV) chamber (base pressure less than $10^{-10}$ Torr). After the deposition of the bottom Fe(001) electrode on both samples, sample A was put into a secondary UHV chamber, adjacent to the growth chamber. Then, molecular O was adsorbed at room temperature on the bottom Fe surface of sample B only. The deposition was stopped after adsorption of one O monolayer. The O adsorption was controlled in real time in situ by x-ray photoelectron spectroscopy (XPS) as described in detail in the previous work [14]. Then, both samples were annealed at 925 K. In sample B, the annealing resulted in $p(1 \times 1)$ ordering of the adsorbed O monolayer. Subsequently the MgO barrier and the top Fe layer were grown. The MgO barrier growth was not affected by the presence of the O layer which was confirmed by observing the RHEED pattern during the barrier deposition [14]. No annealing of the top electrode was performed in order to prevent any further evolution of the bottom interface with respect to the initial configuration. For this reason the second MgO/Fe interface is of lower quality. The most important feature of this procedure is that the thicknesses, growth and annealing conditions for both bottom Fe electrodes and the MgO barrier were strictly identical for both A and B samples. Therefore, the differences in the transport characteristics are only related to the presence of the additional O monolayer at the bottom Fe/MgO interface.

Measurements of the bias dependence of TMR were performed on both Fe/MgO/Fe and Fe/O/MgO/Fe MTJs for 1.1, 1.6 and 2.3 nm MgO barrier thickness. The currents $I_P = I_{↑↑} + I_{↓↓}$ and $I_{AP} = I_{↑↓} + I_{↓↑}$ are measured for magnetizations in the electrodes parallel (P) and antiparallel (AP) to each other. Then the TMR ratio is calculated as $TMR = (I_P - I_{AP})/I_{AP}$. The bias dependence of the TMR in the sample with 1.1 nm of MgO barrier is shown in figure 1. The TMR curves for 1.6 and 2.3 nm are qualitatively similar except for a higher value of TMR and larger noise level. The noise increase with increasing thickness and/or decreasing bias is due to the smaller magnitude of the measured current, which leads to a decrease in precision. The maximum TMR value is obtained at small finite bias from where TMR monotonically decreases with the bias. A similar shift of the maximum to finite bias is observed in most asymmetric MTJs, for example with NiO [9], Fe$_3$O$_4$ [11] or C modified barriers [11, 25].

TMR increases with the thickness of the MgO barrier, with a maximum value of 50% for 1.1 nm, 117% for 1.6 nm and 142% for 2.3 nm for the MTJs with non-oxidized interfaces. The maximum TMR value obtained for the thicker MgO barrier is slightly lower than the ratio measured earlier [25], because in these samples the top Fe electrode has not been annealed and therefore the structural quality of the
MTJ stack is slightly reduced. Comparing samples A and B for the same MgO barrier, we observe that the presence of one monolayer of O at the bottom Fe/MgO interface decreases the TMR in all cases with respect to the O-free samples (to 38% for 1.1 nm, 50% for 1.6 nm, and 115% for 2.3 nm barriers, respectively). These observations are consistent with previous reports [2, 26, 27].

In all cases TMR is asymmetric with respect to the voltage. We also observe that TMR changes sign at large bias from positive to negative. Due to the asymmetry, the inversion happens only at positive bias for the experimental range of voltages, which is limited to prevent electric breakdown. In order to quantify and compare the TMR asymmetry between samples, we define $V_+$ and $V_-$ to be the absolute values of the bias at which TMR reaches half of its maximum value for positive and negative bias, respectively. For the 1.1 nm thick MgO barrier (figure 1) with non-oxidized interfaces we obtain $V_+ = 0.42$ V and $V_- = 1.04$ V. We define an asymmetry coefficient as follows: $\gamma = (V_+ - V_-)/V_- = 60\%$. This asymmetry is large, suggesting that the quality of the two interfaces is markedly different. This is not surprising bearing in mind that the bottom Fe/MgO interface is atomically sharp after the annealing of the bottom Fe electrode, while the top MgO/Fe interface is rougher for a couple of reasons. First, the MgO barrier modifications. In the case of Fe/MgO-based MTJs, this model has predictive power due to the particular band structure of the ferromagnetic leads which can be thought to contain only one exchange-split $\Delta_1$ band [29–36]. This model represents an approximation in a couple of ways: (i) it ignores the contribution to the transport of the bands with symmetries other than $\Delta_1$ and (ii) it does not account for the contribution of the interface resonance state present in the minority Fe. For finite bias the rigid band approximation is used, i.e. that bands are rigidly shifted in energy by the applied bias. The voltage drop is entirely in the barrier region. We assume that the electrostatic potential changes linearly within the barrier. This model has been demonstrated previously to describe remarkably well the behavior, not only of the charge current and TMR behavior [29–31], but also of the spin current and the spin-transfer torque (STT) [32–36]. At the same time the model gives us the opportunity to readily calculate the voltage dependence of the current and TMR in MTJs with a variety of barrier modifications.

The Hamiltonian for the MTJ is

$$H = H_L + H_R + H_S + (H_{SL} + H_{SR} + \text{h.c.})$$

where the different terms represent the isolated left and right leads $H_{L(R)} = \sum \epsilon^\sigma c_i^\dagger \sigma c_i + \sum t_{ab} c_a^\dagger \sigma c^\sigma$, the scattering region $H_S = \sum \epsilon c_i^\sigma c_i^{\dagger \sigma} + \sum t_{ab} f_a f_b^{\dagger}$ and the coupling between the leads with the scattering region $H_{SL(R)} = \sum t_{a\alpha(b\beta)} f_a c_a^{\dagger \alpha} c_{a(\beta)}$. By convention the first principal layers of the left (right) interface are labeled $\alpha(\beta)$ indices in the leads and $a(b)$. 

\[\text{Figure 1. Experimental bias dependence of TMR in single crystal Fe/MgO/Fe and Fe/O/MgO/Fe MTJs. The thickness of the MgO barrier is 1.1 nm. The top Fe electrode is taken as reference while the bias is applied to the bottom Fe electrode. A numerical smoothing procedure is applied to reduce the experimental noise around zero bias.}\]
in the barrier. The spin-split $\Delta_1$ bands in the Fe electrodes are represented by their spin-dependent onsite energies $\varepsilon^{\uparrow} = 3.0$ eV and $\varepsilon^{\downarrow} = 5.6$ eV respectively. The onsite energy in the bulk MgO barrier is $\varepsilon_B = 9.0$ eV (B region). We vary the onsite energies in the B$_L$ and B$_R$ regions to model MTJs with modified interfaces. The nearest neighbor hopping matrix elements are chosen to be $t = -1.0$ eV in all regions. The Fermi energy is at $E_F = 0.0$ eV. The calculations were carried out at low temperature.

The finite bias tunneling current is calculated using the standard Landauer formalism [37, 38]

$$I_{\sigma\sigma'} = e \int dE \frac{d\mathbf{k}_L}{|f(E, \mu_L) - f(E, \mu_R)|} T_{\sigma\sigma'}(E, \mathbf{k}_L)$$

where the transmission probability $T_{\sigma\sigma'}(E, \mathbf{k}_L) = \text{Tr}[\Gamma_L^\sigma \Gamma_R^{\sigma'} | \Gamma_R^\sigma \Gamma_L^{\sigma'} ]$ is integrated over $k_L$ in the surface Brillouin zone and over energy within the bias window. The bias window is controlled by the Fermi–Dirac distribution $f(k_L)$.

Similarly, we calculate the bias dependence of the TMR in Fe/MgO/Fe and FeO/MgO/Fe MTJs. Oxygen in FeO/MgO/Fe MTJs is represented by 1 monolayer at the left interface with onsite energy $\varepsilon_{B_L}$. The rough right interface is represented with 1 monolayer with onsite energy $\varepsilon_{B_R}$. The thickness of the middle part of the MgO barrier is 4 monolayers.

The transmission probability at the Fermi energy is at $E_F = 0.0$ eV, which still represents a barrier layer albeit smaller than the bulk. The calculated TMR as a function of the bias and at the Fermi point only, it reduces to the familiar Julliere formula $\text{TMR} = 2P_L P_R/(1 - P_L P_R)$, where $P_L(R) = (t_{L(R)}^\uparrow - t_{L(R)}^\downarrow)/(t_{L(R)}^\uparrow + t_{L(R)}^\downarrow)$ is the spin polarization of the DOS induced in the barrier by the electrodes.

Using this model we calculate the tunneling current and TMR in Fe/MgO/Fe MTJs corresponding to the experimental setup. For sample A we assume that at the sharp interface the barrier is identical to the bulk $\varepsilon_{B_L} = \varepsilon_B = 9.0$ eV, while at the rough interface the barrier height is reduced. We vary $6.0 < \varepsilon_{B_L} < 9.0$ eV, which still represents a barrier layer albeit smaller than the bulk. The calculated TMR as a function of the applied bias is shown in figure 2. It can be seen that when $\varepsilon_{B_L}$ is close to the bulk value, the TMR is symmetric and close to the ideal case. As $\varepsilon_{B_L}$ decreases, the maximum value of TMR decreases and the asymmetry is increased. Moreover, we see that for large enough bias TMR changes sign, but due to the asymmetry this inversion happens for much smaller bias in one direction than in the other. These results show qualitative agreement with the experimental observations. An asymmetry coefficient approximately equal to the experimental one is obtained for the value of $\varepsilon_{B_L} = 7.0$ eV. Thus, this model allows us to interpret many similar experiments in which one of the interfaces is morphologically different [16–19]. In those cases, disorder and roughness make the barrier at the interface more diffuse, effectively lowering the barrier height, which gives rise to asymmetric behavior.

Similarly, we calculate the bias dependence of the TMR in sample B, where we fix $\varepsilon_{B_L} = 7.0$ eV to account for the rough interface and vary the onsite energy of the O monolayer $7.0 < \varepsilon_{B_R} < 9.0$ eV. The results, plotted in figure 2, show that the asymmetry is reduced. The same asymmetry as in experiment ($\gamma = 28\%$ for 1.1 nm of MgO barrier) is obtained for $\varepsilon_{B_R} = 7.5$ eV. Thus, within our model, the O layer can be
understood as an additional barrier. This agrees well with the experimental observation that the O layer attenuates both the P and AP spin channels the same, which behavior is consistent with an additional barrier at the interface [14].

In the case when all parts of the barrier are insulating, the ITFs \(t_L\) and \(t_R\) are largely determined by the DOS at the electrodes. Thus, the decrease in TMR and the subsequent sign reversal at finite bias, for both symmetric and asymmetric MTJs, could be qualitatively explained in terms of the realignment of majority- and minority-spin DOS in the left and right electrodes [8]. The band alignments at zero and finite basis are illustrated in figure 3(a). At zero bias the majority and minority bands are aligned in the P configuration, and misaligned in the AP configuration (due to exchange splitting). The propagating states in the electrodes in the P configuration experience no scattering, while in the AP configuration the majority and minority bands are aligned in the P configuration and misaligned in the AP configuration (due to exchange splitting). The propagating states in the electrodes in the P configuration experience no scattering, while in the AP a large portion of the states is scattered. This happens when the bias is comparable to the exchange splitting, aligning the majority and minority bands, in which case the transmission probability is dominated by one of the channels in the AP configuration \((I_{↑↑} \text{ for negative bias or } I_{↑↓} \text{ for positive bias})\).

For symmetric MTJs the bands in the P orientation at zero bias have an optimum alignment and no scattering. Thus any bias introduces some scattering and causes \(I_P\) to increase with bias slower than \(I_{AP}\). In asymmetric MTJs, however, the ITFs at the two interfaces are not equal. The potential profiles of a barrier with an insulating layer of lower barrier height at the right interface are drawn in figure 3(b) for zero and finite bias. In this case \(t_R > t_L\) and there is some scattering in the P channel even at zero bias. One of the effects of this is that TMR is lowered compared to the symmetric case because the P channel conduction is suboptimal. Applying negative bias decreases the effective barrier height at the left interface and increases it at the right interface. This will tend to decrease \(t_R\) and increase \(t_L\), reducing the scattering in the P channel and making \(I_P\) increase faster than \(I_{AP}\). Positive bias works in the opposite direction. Thus, the second effect is that the TMR maximum switches to finite negative bias.

When the bias becomes comparable to the size of the barrier reduction at the right interface \(I_{AP}\) will again outpace \(I_P\). However, since negative bias in general improves the alignment and reduces the scattering in the P configuration it will take larger negative bias, compared to the symmetric barrier, before \(I_{AP} > I_P\) and TMR becomes negative. Positive bias does the opposite and smaller positive voltage is required to change the sign of TMR.

These observations allow us to generalize the model to explain experiments in which extra insulating layers are inserted at the interface, in particular NiFe/Fe\(_2\)O\(_3\)/Al\(_2\)O\(_3\)/NiFe [8] and CoFe/NiO/MgO/CoFe MTJs [10]. In both cases, asymmetric bias dependence of TMR and TMR sign inversion at high bias were observed. For example, to model the NiO/MgO composite barrier, we choose \(\varepsilon_{B R} = 9.0\ \text{eV}\), the same as the bulk, and \(\varepsilon_{B L} = 7.0\ \text{eV}\), because the band gap of NiO is smaller compared to MgO. The calculated bias dependence of TMR is shown in figure 4. TMR is symmetric for unmodified MgO barriers. The insertion of a thin NiO layer at the interface introduces asymmetry in the bias dependence of TMR. Thus, for positive (negative) bias higher (lower) voltage is required to change the sign of TMR. However, placing the NiO layer inside the MgO barrier essentially restores the symmetry in the bias dependence of TMR. When the NiO layer is at the interface \(t_L\) is different from \(t_R\) resulting in asymmetric bias dependence of TMR. However a slight shift of the NiO layer from the interface yields \(t_L \approx t_R\) and almost negligible asymmetry of TMR comes only through the asymmetric weighting with the S-matrix in the integral. Thus, the observed bias dependence and inversion of TMR can be understood simply in terms of asymmetric ITFs and the exchange bias in the electrodes.

Further on, the model can be used to explain the bias dependence of TMR in MTJs with metallic interface layers, such as Fe/Cr/MgO/Fe [10] and Fe/Fe\(_3\)O\(_4\)/MgO/CoMTJs [11]. We model metallic layers by lowering the onsite energy of the interface layer \(\varepsilon_{B L} < 6.0\ \text{eV}\) in which case the band crosses the Fermi level. The results for TMR are shown in figure 5. 

![Figure 3](image-url)
for several voltages. In the tunneling regime TMR ($\varepsilon_{R_L} = 6.0$ eV) has the ‘usual’ form, with a maximum close to zero voltage and behaving as an even function of the voltage. For a shallow potential well at the interface ($\varepsilon_{R_L} = 5.0$ eV) the bias dependence of TMR already shows qualitatively different behavior; TMR inversion cannot be achieved by applying positive voltage. In this case we notice the interesting feature that TMR becomes an odd function of the bias. This suggests a possibility of combining memory and logic functions in the same bit [43, 44]. Moreover, TMR increases with voltage. In general memory stacks are operated at low voltage because TMR has a maximum. This feature can allow memories to be operated at higher voltage. Similar behavior but with reverse sign is observed for $\varepsilon_{R_L} = 4.0$ eV the minority resonance starts to overlap with the electrode DOS, which results in very strong AP transmission, $I_{AP} > I_P$, and TMR becomes negative. As $\varepsilon_{R_L}$ increases the resonances disappear altogether and we have a transition to the insulating regime at $\varepsilon_{R_L} = 6.0$ eV. Conversely for lower $\varepsilon_{R_L}$ or for thicker interface more than one resonance can appear.

4. Conclusions

We report asymmetric bias dependence of TMR and TMR reversal at large bias in single crystal Fe/MgO-based MTJs: one with two morphologically different interfaces and one with a layer of O inserted at the bottom interface. We develop a model to describe the TMR behavior in MTJs with modified barriers, with the added benefit that it allows us to reinterpret and categorize the wide variety of experimental results. The model, although simple, captures the essential physics of the Fe/MgO-based MTJs (namely the $\Delta_1$ filtering) and agrees qualitatively with the experimental data. It predicts two distinct regimes: tunneling when the modified interfaces are insulating, and resonant when the interfaces become metallic. In the tunneling regime, we observe asymmetric bias dependence of the TMR resulting from the asymmetry of the ITFs due to the difference of the morphology of the interfaces. We explain the TMR inversion results from the realignment of the electrode DOS in large bias, which overcomes the exchange splitting in the electrodes. In the resonant regime, the electron tunneling through the resonant levels at the interface can dominate the transmission probability for a particular spin channel. Judicious choices of the position of the resonant level allow for control of the shape of the TMR bias dependence. We have demonstrated cases of normal TMR decreasing with bias, inverse TMR increasing with bias and TMR changing sign for positive and negative bias.
Figure 6. $k_{\parallel}$-resolved majority and minority DOS in the electrodes (left panels), and induced DOS in the first barrier layer at the left interface for $\varepsilon_{BL1} = 3$ eV (middle panels) and $\varepsilon_{BL1} = 4$ eV (right panels).

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