Theory of tunneling magnetoresistance in a disordered Fe/MgO/Fe(001) junction

How to cite:

For guidance on citations see FAQs

© 2006 The American Physical Society
Version: Version of Record
Link(s) to article on publisher's website:
http://dx.doi.org/10.1103/PhysRevB.74.140404

Copyright and Moral Rights for the articles on this site are retained by the individual authors and/or other copyright owners. For more information on Open Research Online’s data policy on reuse of materials please consult the policies page.

oro.open.ac.uk
Theory of tunneling magnetoresistance in a disordered Fe/MgO/Fe(001) junction

J. Mathon\textsuperscript{1} and A. Umerski\textsuperscript{2}

\textsuperscript{1}Department of Mathematics, City University, London EC1V 0HB, United Kingdom
\textsuperscript{2}Department of Applied Mathematics, Open University, Milton Keynes MK7 6AA, United Kingdom

(Received 5 July 2006; revised manuscript received 9 August 2006; published 20 October 2006)

Calculation of the tunneling magnetoresistance (TMR) of an Fe/MgO/Fe(001) junction with a disordered Fe/MgO interface is reported. It is shown that intermixing of Fe and Mg atoms at the interface decreases the TMR ratio rapidly and when about 16% of interfacial Fe atoms are substituted by Mg the calculated TMR saturates with increasing MgO thickness in good agreement with experiment. It is demonstrated that the saturation of TMR occurs because interfacial scattering leads to a redistribution of conductance channels, which opens up the perpendicular tunneling channel in the antiferromagnetic configuration that is forbidden for a perfect epitaxial junction.

DOI: 10.1103/PhysRevB.74.140404

PACS number(s): 75.70.Cn, 75.45.+j

It was predicted theoretically\textsuperscript{1,2} that a very large tunneling magnetoresistance (TMR), in excess of 1000\%, can be achieved for an epitaxial Fe/MgO/Fe(001) tunneling junction. Although recent measurements for this system\textsuperscript{3–5} give very large TMR ratios of the order of 300\%, the situation remains somewhat controversial since there is one major aspect in which the theoretical results for a perfect Fe/MgO/Fe(001) junction differ fundamentally from the observed results. The theory predicts that the TMR ratio increases with increasing MgO thickness and there is no theoretical limit on the magnitude of TMR that can be achieved for a perfect epitaxial junction with a very thick MgO barrier. The calculated\textsuperscript{1} dependence of the optimistic TMR ratio on MgO thickness is reproduced in Fig. 1(a) (broken curve). The TMR ratio increases with MgO thickness because symmetry impedes perpendicular tunneling of minority-spin electrons at the \( \Gamma \) point of the two-dimensional (2D) Brillouin zone.\textsuperscript{2} When the tunneling conductance in the antiferromagnetic (AF) configuration is plotted in the 2D Brillouin zone it exhibits\textsuperscript{1} a “hole” around the \( \Gamma \) point. This is illustrated in Fig. 1(b). It follows that the tunneling conductance in the AF configuration decays asymptotically exponentially with increasing MgO thickness at a rate higher than that in the ferromagnetic (FM) configuration in which tunneling of majority-spin electrons at the \( \Gamma \) point is allowed.

In contrast to these theoretical predictions, the experimental results of Yuasa \textit{et al.},\textsuperscript{3} which are also reproduced in Fig. 1 (circles), show a rapid saturation of TMR with increasing MgO thickness. It is essential to identify the physical mechanism that controls the observed saturation of TMR since we can then try to manipulate it to reach the desired theoretical regime in which TMR increases asymptotically monotonically with MgO thickness.

We propose that the physical mechanism that causes the saturation of TMR is a relatively small amount of disorder at one (or both) of the Fe/MgO interfaces. In particular, we shall show that intermixing of Fe and Mg atoms at one of the Fe/MgO interfaces can reproduce very well the observed behavior of TMR as a function of MgO thickness. Such intermixing of Fe and Mg atoms certainly takes place if a small amount of FeO is formed at the interface.\textsuperscript{6} The fact that the measured conductances\textsuperscript{3} are almost perfect straight lines when plotted against MgO thickness on a logarithmic scale suggests that the MgO barrier itself is of a high quality. Even if there are imperfections in the bulk of MgO, it will be seen that they cannot explain the observed saturation of TMR. We shall therefore assume in our calculations that the MgO barrier is perfect.

The method of choice for investigating the effect of interfacial intermixing is a lateral supercell geometry. While the method is straightforward in principle, one encounters serious numerical problems when applying it to a disordered Fe/MgO/Fe(001) junction. The aim is to evaluate the conductances \( \tilde{\sigma}_\sigma \) of electrons of spin \( \sigma \) tunneling through a disordered junction from the Kubo-Landauer formula\textsuperscript{1} in a supercell geometry. As before,\textsuperscript{1} we use tight-binding bands fitted to an \textit{ab initio} band structure of Fe and the barrier is described by tight-binding bands fitted to the band structure of bulk MgO.\textsuperscript{7} For details of our tight-binding parametrization of the Fe/MgO/Fe(001) junction see Ref. 1. Previously,\textsuperscript{1} we fixed the position of the common Fermi level \( E_F \) in the middle of the MgO gap of 7.6 eV. This is correct for a perfect epitaxial junction.\textsuperscript{8,9} However, when an interfacial intermixing is present metal-insulator gap states (MIGS) may form at the interface, and such states determine ultimately the position of \( E_F \) in the MgO gap.\textsuperscript{10} Without detailed knowledge of the nature of the MIGS, the position of \( E_F \) in the MgO gap cannot be determined from first principles. We therefore treat the position of \( E_F \) in the MgO gap as the only

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1}
\caption{(a) The computed TMR for a perfect epitaxial junction (broken curve) and the observed TMR (Ref. 3) (circles); (b) distribution in the two-dimensional Brillouin zone of the partial conductances in the antiferromagnetic configuration of the perfect epitaxial junction.}
\end{figure}
lateral supercell geometry takes the form
parameters within the lateral cell. The Kubo formula in the
layers, plane index $i$, the values of all the decay constants (effective
heights of the gap) can be determined from the complex
Fermi surface of MgO. 1

We use a mixed representation that is Bloch-like in the
direction parallel to the layers and atomic-like in the perpen-
dicular direction. We shall label all quantities in the supercell
geometry with a tilde. The one-electron Green’s functions $\tilde{G}$
at the Fermi surface $E=E_F$, which are required in the Kubo
formula, depend on the supercell wave vector $\tilde{k}_i$ parallel to
the layers, plane index $i$, and indices $r,s$ labeling atomic
positions within the lateral cell. The Kubo formula in the
lateral supercell geometry takes the form
\[
\tilde{\Gamma}^{\sigma} = \frac{4e^2}{\hbar} \sum_{k} \text{Tr}[\tilde{T}_\sigma \text{Im} \tilde{G}_r^{\sigma}(\tilde{k}_i,r,r) \cdot \tilde{T}_\sigma \text{Im} \tilde{G}_r^{\sigma}(\tilde{k}_i,r,r)] .
\]  

The summation in Eq. (1) is over the supercell two-
dimensional Brillouin zone and the cell index $r$. The trace is
over the orbital indices corresponding to $s,p,d$ orbitals that
are required in a tight-binding parametrization of the junc-
tion. Finally, $\tilde{G}_r^{\sigma}(\tilde{k}_i,r,s)$ and $\tilde{G}_r^{\sigma}(\tilde{k}_i,r,s)$ are the one-electron
Green’s functions at the left (right) surfaces of a junction that
is separated into two independent parts by an imaginary
cleavage plane drawn between any two neighboring atomic
planes $L,R$. Naturally, the total tunneling current in each spin
channel is conserved and, therefore, it is immaterial in the
calculation of TMR where the cleavage plane is positioned.
However, it will be seen that for a physical interpretation of
our results, it is convenient to place the cleavage plane in
two alternative positions. These are denoted by $A$ or $B$ in Fig.
2, which shows schematically the junction we consider. We
place the cleavage plane either immediately to the left of an
interfacial atomic plane in which intermixing between Fe and
Mg atoms is assumed to occur ($L=0$, $R=1$) or immedi-
ately to the right of this plane ($L=1$, $R=2$). The interface
between MgO and the right Fe electrode is assumed to be
perfect.

The separation of the junction into two independent parts
is made simply for calculational purposes. The junction re-
mains physically connected and the interaction between the
left and right parts is fully restored in Eq. (1) by the matrices
$\tilde{T}_\sigma$ and $\tilde{T}_\sigma$ defined by
\[
\tilde{T}_\sigma = t_{LR}(\tilde{k}_i)[\tilde{I} - \tilde{G}_r^{\sigma}(\tilde{k}_i)\tilde{G}_r^{\sigma}(\tilde{k}_i)]^{-1} ,
\]
where $\tilde{I}$ is a unit matrix and $t_{LR}(\tilde{k}_i)$ is the tight-binding hopping
matrix connecting the surfaces $L$ and $R$.

All the matrices in Eqs. (1) and (2) operate in a vector
space that is a direct product of the space of atomic orbitals
and the space of supercell indices $r,s$. For a typical $10 \times 10$
supercell, which we use here, the dimensions of all the ma-
trices are $900 \times 900$ since nine orbitals are used in our tight-
binding parametrization. We recall that even calculations of
the tunneling conductances for a perfect Fe/MgO/Fe(001)
junction are numerically highly demanding since a very
small imaginary part to the energy, of the order of $10^{-12}$ Ry,
must be combined with a very fine mesh of $k_1$ points to
achieve convergence. We had to use in our original
calculations up to $10^8 k_1$ points in the irreducible segment
of the 2D Brillouin zone (BZ). When we tried to implement
this program for a supercell with $900 \times 900$ matrices, the
problem became numerically unstable. We have, therefore,
developed an alternative strategy that not only delivers the
required numerical accuracy but also gives a very clear
physical interpretation of our supercell calculations.

To evaluate the Kubo formula, we first determined the
surface Green’s function $G_{r,r}(\tilde{k}_i)$ for the left Fe electrode
(plane 0 in Fig. 2) and the surface Green’s function $G_{r,r}(\tilde{k}_i)$
for the left surface of MgO deposited on the right Fe elec-
trode (plane 2 in Fig. 2) using the simple cell basis. That
involves only operations with matrices of the same small size
as for a perfectly epitaxial junction. Only in the last step of
“depositing” the mixed layer and connecting the left and
right surfaces across the cleavage plane via Eq. (2) do we
convert to the supercell basis. Such a conversion is possible
since the simple and supercell bases span the same Hilbert
space and there is, therefore, a unitary transformation $\tilde{M}$
$=UMU^{-1}$ that transforms any matrix $M$ with matrix elements
$M(k_1,k')$ in the simple cell basis to a matrix $\tilde{M}$ with matrix
elements $\tilde{M}(\tilde{k}_1,\tilde{k}_1',r,s)$ in the supercell basis, and vice versa
(suppress the orbital and plane indices since they remain
the same in the simple and supercell representations). This
method renders the calculation of the TMR for a disordered
junction feasible.

To determine the TMR, we first calculated from Eqs. (1)
and (2) the FM and AF tunneling conductances for different
amounts of intermixing and performed a configuration aver-
age over 30 random configurations of Mg impurities. All the
conductances we obtained are perfect straight lines when
plotted on a logarithmic scale as a function of MgO thick-
ness. Moreover, the tunneling conductance in the FM con-
figuration $\tilde{\Gamma}_{FM}$ is quite insensitive to the amount of intermix-
ing. The reason for this behavior will be explained later. We
have, therefore, compared the slope of the calculated and
observed conductances $\tilde{\Gamma}_{FM}$ to determine the position of $E_F$. The
best fit is obtained for $E_F$ lying about 2 eV above the top
of the valence band of MgO. This value also represents the

FIG. 2. Fe/MgO/Fe(001) junction with intermixing of Fe and
Mg atoms at the left Fe/MgO interface; the two alternative posi-
tions of a cleavage plane are denoted by $A$ and $B$. 
FIG. 3. The calculated TMR ratio for different amounts of intermixing between Fe and Mg atoms and the observed TMR ratio (circles).

effective height of a barrier for majority-spin electrons in the FM configuration. Using this value, we have calculated the TMR ratio for different amounts of intermixing between Fe and Mg atoms. The calculated results are compared in Fig. 3 with the experimental results of Yuasa et al.\(^1\) Since the TMR can only be calculated for thicknesses of MgO corresponding to complete atomic planes, we show in Fig. 3 the experimental results also for such discrete thicknesses (circles).

It can be seen that intermixing decreases the TMR ratio rapidly, and when some 16% of interfacial Fe atoms are substituted by Mg the calculated TMR saturates with MgO thickness in good agreement with the experiment.\(^3\)

We now clarify the physical mechanism that causes the saturation. We propose that it is a redistribution of the partial conductances \(\Gamma^{\sigma}(k_{1},k_{3})\) on \(k_{1}\) for the position \(B\) of the cleavage plane. The thickness of the MgO barrier is ten atomic planes.

[TM image showing distribution of partial conductances]

FIG. 4. Distribution of the partial conductances in the two-dimensional Brillouin zone for the position \(B\) of the cleavage plane: (a) \(\Gamma_{FM}^{↑}\), (b) \(\Gamma_{AF}^{↑}\), (c) \(\Gamma_{AF}^{↑}\), (d) \(\Gamma_{AF}^{↓\downarrow}\).
electrode and can thus propagate at the $\Gamma$ point across the whole junction. Therefore, the hole at the $\Gamma$ point is removed. This demonstrates explicitly our argument that interface roughness removes the hole at the $\Gamma$ point in the AF configuration. It should be noted that the “lattice” structure appearing in Figs. 5(b) and 5(d) is due to the finite size of our supercell. For a very large supercell, electrons from the whole cross section of the minority-spin surface spectral density of Fe(001) would contribute to tunneling.

It is appropriate to mention here that imperfections in the bulk of MgO cannot explain the saturation effect. This is because by the time $\downarrow$-spin carriers emitted from Fe at the $\Gamma$ point reach an impurity in the bulk of MgO their wave functions have already decayed rapidly and, hence, the $\Gamma$ point $\downarrow$-spin conductance channel remains blocked. It is essential that scattering take place at (or very near) the interface so that $\downarrow$-spin carriers are scattered toward the $\Gamma$ point just when they enter MgO [Fig. 5(d)] and can thus tunnel across the whole thickness of MgO with the same decay constant as $\uparrow$-spin carriers.

Finally, we shall briefly discuss the observed small oscillations of TMR with MgO thickness. Such oscillations could be fundamental since the complex Fermi surface of MgO has both imaginary and real sheets. The real part of the perpendicular wave vector in MgO could give rise to oscillations of TMR. However, since the MgO complex Fermi surface has no real part in a large region around the $\Gamma$ point, this cannot be the correct explanation of the observed oscillations. Furthermore, the fact that the observed oscillations do not decrease with increasing MgO thickness is further evidence that they do not arise from the MgO complex Fermi surface. On the other hand, it is clear that the amount of interfacial roughness varies periodically with MgO thickness and is smallest for thicknesses corresponding to most perfect interfaces. Since we have demonstrated that the magnitude of TMR depends critically on interfacial roughness, growth-induced oscillations of TMR with a period of approximately one atomic plane must occur. However, since the period of the observed oscillations is somewhat longer their precise origin requires further investigation.

We have demonstrated that a small amount of intermixing of Fe and Mg atoms at one of the Fe/MgO interfaces can explain the observed saturation of TMR with MgO thickness. It could be argued that one should also include in the rough interface an admixture of oxygen atoms to simulate small random regions of FeO. This would be desirable, and is possible in principle, but it would require very large lateral supercells, which would make reliable calculations of TMR much more difficult. However, it is clear from our discussion (see Figs. 4 and 5) that it is the existence of a disorder, rather than any particular type of disorder, that causes the saturation effect. It follows that other types of imperfection such as dislocations or oxygen vacancies would have qualitatively the same effect. Disorder at both interfaces gives qualitatively similar results but TMR saturates more rapidly.

Finally, the position of the Fermi level about 2 eV above the top of the valence band of MgO, which we deduced by comparing the calculated and measured tunneling conductances, is consistent with the height of the barrier quoted by Parkin et al. and with the weak bias dependence of TMR reported by Yuasa et al.