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Asymptotic behavior of tunneling magnetoresistance in epitaxial MgO tunnel junctions

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Almost a decade after the theoretical prediction of large tunneling magnetoresistance ratio (TMR) in epitaxial Fe/MgO/Fe tunneling junctions, there is still debate on how the TMR should behave as a function of MgO thickness for thick MgO. In this Brief Report we prove that the optimistic TMR grows indefinitely with MgO thickness, and in particular, that for large MgO thicknesses it must grow linearly. This rather surprising result is obtained using straightforward and completely general asymptotic analysis and is applicable to other tunneling junctions with MgO barrier. We give a simple formula for the growth of TMR and show that it is in excellent agreement with computational studies on realistic epitaxial systems. The formula also provides a valuable test to check that numerical studies are converging to the correct answer.

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The original theoretical prediction[1,2] and subsequent observation[3,4] of a very large tunneling magnetoresistance ratio (TMR) in epitaxial junctions with crystalline MgO barrier and Fe (or Co) electrodes, has lead to intense study of these systems in recent years. Most of the theoretical study has been concerned with the nature of tunneling through an MgO barrier when the barrier is relatively thin, i.e., of the order of 2–8 atomic planes. Relatively little theoretical attention has been given to the behavior of the optimistic TMR for thicker barriers. Note that the optimistic TMR is defined by

$$\mathrm{TMR} = \frac{\mathcal{G}_{\mathrm{P}} - \mathcal{G}_{\mathrm{AP}}}{\mathcal{G}_{\mathrm{AP}}}, \tag{1}$$

where $\mathcal{G}_{\mathrm{P}}$ and $\mathcal{G}_{\mathrm{AP}}$ are the total conductances in the parallel (P) and antiparallel (AP) configurations of the magnetic leads.

Most experimental data[3,5] appears to show that the optimistic TMR initially grows quickly with MgO thickness, then saturates at above 8–10 atomic planes of MgO. The saturation value however depends on factors such as annealing temperature,[5] which most likely affects the quality of the Fe/MgO interface. More recent results[6] seem to suggest that the optimistic TMR grows approximately linearly with MgO thickness up to 15 atomic planes, the largest thickness measured in these experiments. Although one should be aware that we have based this inference on just three data points taken from Fig. 2 of Ref. 6.

Most theoretical results do not display the optimistic TMR as a function of MgO thickness. This is unfortunate as it is crucial to make contact with experiment. It was shown by Butler *et al.*[2] that a very large TMR for a perfect epitaxial junction is expected because of special features of the Fe/MgO band structure at the $\bar{\Gamma}$ point (perpendicular tunneling). These ensure that the conductance G_{P} at the $\bar{\Gamma}$ point has a slower exponential decay with MgO thickness than G_{AP}. Furthermore, as the thickness of MgO increases, tunneling is progressively restricted toward the $\bar{\Gamma}$ point and one might therefore expect that the optimistic TMR should grow exponentially with MgO thickness. However this conclusion is incorrect. This strict suppression of minority spin electron tunneling is only valid at the $\bar{\Gamma}$ point, and as one moves away from this point, the decay rate of the conductance G_{AP} becomes the same as that of G_{P}. On the basis of this argument Heiliger *et al.*[8] concluded that the optimistic TMR for a perfect epitaxial MgO-based junction must eventually saturate with MgO thickness. This conclusion is also incorrect. Instead there is a subtle competition between the above process and the fact that the tunneling regime shrinks toward the $\bar{\Gamma}$ point as the barrier thickness increases. Qualitative arguments are not sufficient to determine the outcome of this competition. The purpose of this Brief Report is to give a rigorous mathematical argument, based on completely general asymptotic analysis, which determines the exact asymptotic dependence of TMR on MgO thickness. Surprisingly this rigorous argument shows that the optimistic TMR neither grows exponentially nor saturates but rather it grows linearly with MgO thickness. We give a simple formula for this growth and show that it is in exact agreement with previously published fully numerical calculations of Mathon and Umerski.[7] It is interesting to note that despite the incorrect conclusion of Heiliger *et al.*,[8] their computed results (see Fig. 2 of Ref. 8) also support a linear dependence rather than any saturation.

This linear dependence is valid only for a strictly epitaxial MgO-based junction. The experimentally observed saturation of the TMR is thus not an intrinsic property of a perfect junction as Heiliger *et al.*[8] claim but can only occur as a result of disorder. This was discussed in detail by Mathon and Umerski[7] and their calculations show that disorder on the Fe/MgO interface causes eventual saturation of the optimistic TMR with MgO thickness, in almost perfect agreement with experimental studies.[3]

We now proceed to derive the asymptotic formula for the dependence of TMR on MgO thickness. We consider an epitaxial MgO based tunneling junction such as Fe/MgO/Fe (see Fig. 1). The two in-plane directions (x and y) are generically labeled $\|$ and the out of plane direction labeled z.

Figure 2 shows the imaginary Fermi surface of MgO with the imaginary Fermi sheets labeled $k_1, k_2, \ldots$. Note that there

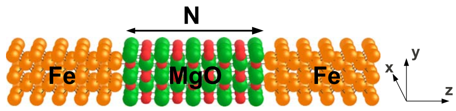

FIG. 1. (Color online) Geometry of an MgO junction.

are also parts of the Fermi surface which have both imaginary and real parts. These real parts are not displayed in Fig. 2. They occur some distance away from the $\bar{\Gamma}$ point (in the region $|k_x| \gtrsim 0.6\pi$ in Fig. 2) and so do not contribute to the asymptotic dependence of TMR.[11]

The meaning of the imaginary sheets in Fig. 2 is that electrons with in plane momentum $\mathbf{k}_\parallel$ must have a wave function of the form $\psi(\mathbf{k}_\parallel) \sim \exp[\pm k_i(\mathbf{k}_\parallel)z]$ and hence tunnel through MgO with decay constant k_i. From this, it can easily be shown that, for a given $\mathbf{k}_\parallel$ the conductance for the system depicted in Fig. 1 must have the form

$$G(\mathbf{k}_\parallel) \sim \sum_i A_i(\mathbf{k}_\parallel)\exp[-2k_i(\mathbf{k}_\parallel)N], \tag{2}$$

where N is the thickness of the MgO junction and the A_i are terms which are independent of N.

In the region around the $\bar{\Gamma}$ point, the lowest lying Fermi sheet is labeled k_1 and hence for thick MgO, electrons preferentially tunnel through this band and with in-plane momentum $\mathbf{k}_\parallel \sim 0$. The mechanism by which one obtains a very large TMR in an Fe/MgO/Fe type junction is now well understood.[1,2] At the Fermi level, the majority spin band in Fe and the k_1 band in MgO both have Δ_1 symmetry at the $\bar{\Gamma}$ point. On the other hand, the minority spin band in Fe does not have this symmetry. It follows that when the Fe leads have parallel magnetization, majority spin electrons can tunnel through the k_1 band at the $\bar{\Gamma}$ point, whereas minority spin electrons cannot, they must tunnel through other bands such as k_2. In the antiparallel configuration, majority electrons in one lead are minority electrons in the other lead and vice versa, so that no electrons can tunnel through the k_1 band at the $\bar{\Gamma}$ point, i.e., they must all tunnel through less preferential bands. Away from the $\bar{\Gamma}$ point, the conducting states both in the ferromagnetic leads and in MgO acquire other symmetries and these strict filtering rules do not apply. As one moves further away from the $\bar{\Gamma}$ point, electrons in the anti-

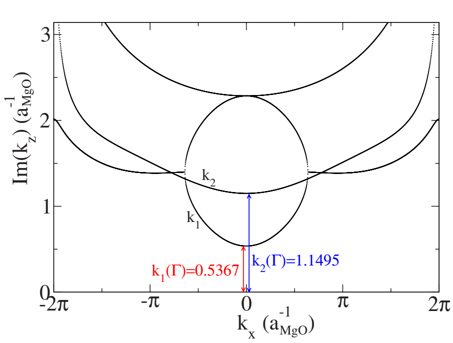

FIG. 2. (Color online) Imaginary Fermi surface of MgO along $\mathbf{k}_\parallel = k_x$.

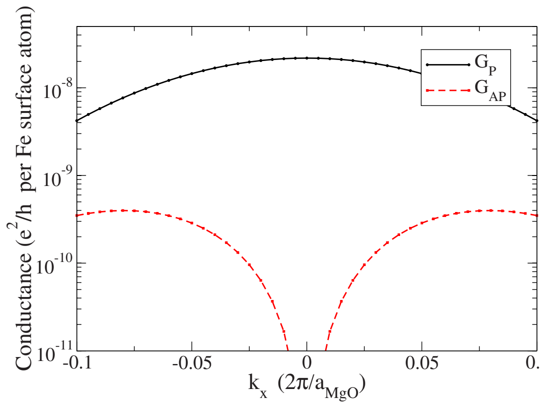

FIG. 3. (Color online) Conductances in the parallel and antiparallel configurations along $\mathbf{k}_\parallel = k_x$, for $N=15$ atomic planes of MgO.

parallel configuration increasingly flow through k_1. Figure 3 depicts the conductance in the parallel and antiparallel configurations, in the region around the $\bar{\Gamma}$ point, for an MgO thickness of $N=15$ atomic planes.

In the parallel configuration the conductance has the form

$$G_\mathrm{P}(\mathbf{k}_\parallel) = A_\mathrm{P}(\mathbf{k}_\parallel)\exp[-2k_1(\mathbf{k}_\parallel)N] + \cdots, \tag{3}$$

where the $\cdots$ indicates terms which decay faster in N than the leading term. The amplitude, $A_\mathrm{P}(\mathbf{k}_\parallel)$ does not vanish at the $\bar{\Gamma}$ point, and so in a neighborhood of the $\bar{\Gamma}$ point it has the form

$$A_\mathrm{P}(\mathbf{k}_\parallel) = A_0 + A_2\mathbf{k}_\parallel^2 + O(\mathbf{k}_\parallel^4). \tag{4}$$

In the antiparallel configuration the conductance must have the form

$$G_\mathrm{AP}(\mathbf{k}_\parallel) = B_\mathrm{AP}(\mathbf{k}_\parallel)\exp[-2k_1(\mathbf{k}_\parallel)N] + C_\mathrm{AP}(\mathbf{k}_\parallel)\exp[-2k_2(\mathbf{k}_\parallel)N] + \cdots. \tag{5}$$

Note that in this case we include the first two leading terms because $B_\mathrm{AP}(\bar{\Gamma})=0$, whereas $C_\mathrm{AP}(\bar{\Gamma}) \neq 0$, indicating that electrons in the antiparallel configuration do not tunnel through the k_1 band at the $\bar{\Gamma}$ point. In a neighborhood of the $\bar{\Gamma}$ point B_AP must have the form

$$B_\mathrm{AP}(\mathbf{k}_\parallel) = B_2\mathbf{k}_\parallel^2 + B_4(k_x^4 + k_y^4) + O(\mathbf{k}_\parallel^6). \tag{6}$$

Note that in principle A_P, and B_AP only have the cubic symmetry of the lattice. However we find to a very high degree of accuracy, in the region of the $\bar{\Gamma}$ point up to order $\mathbf{k}_\parallel^2$, that they are circularly symmetric and so Eqs. (4) and (6) are quite adequate. We do find that at $O(\mathbf{k}_\parallel^4)$ the circular symmetry is broken, and hence the non circularly symmetric term coupled to B_4. Note that the other cubic symmetry term $k_x^2k_y^2$ at $O(\mathbf{k}_\parallel^4)$ is relatively unimportant in our final formula for the TMR and so, for simplicity, we omit it.

The total conductance is obtained by summing all conductance channels over the two-dimensional (2D) Brillouin zone (BZ)

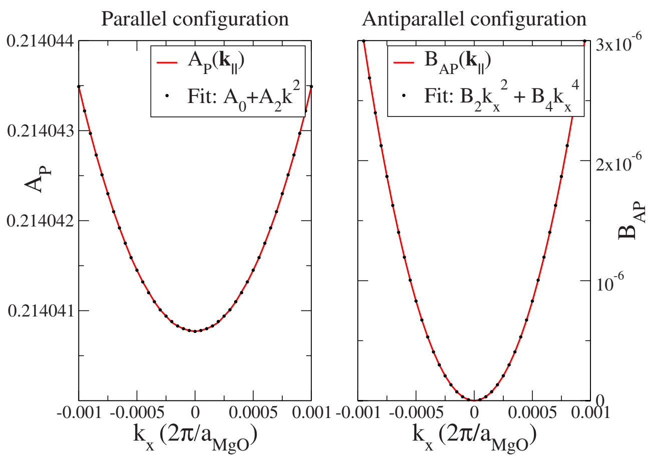

FIG. 4. (Color online) Amplitudes $A(\mathbf{k}_\parallel)$ and $B(\mathbf{k}_\parallel)$ along $\mathbf{k}_\parallel = k_x$, in a region around the $\bar{\Gamma}$ point.

$$\mathcal{G} = \int_{BZ} d\mathbf{k}_\parallel G(\mathbf{k}_\parallel). \tag{7}$$

Since the conductances have the form given in Eqs. (3) and (5), the main contribution to the integral comes from the minimum of k_1 and k_2 which occur at $\mathbf{k}_\parallel = 0$ (see Fig. 2). If we expand $k_i(\mathbf{k}_\parallel)$ about $\mathbf{k}_\parallel = 0$ to second order, in a basis in which the components of $\mathbf{k}_\parallel$ are aligned along the principal axes, then the Hessian matrix of k_i is proportional to the unit matrix, and so we obtain

$$k_i(\mathbf{k}_\parallel) = k_i^0 + \frac{1}{2}H_i\mathbf{k}_\parallel^2$$

where

$$H_i = \left.\frac{\partial^2 k_i}{\partial k_x^2}\right|_{\mathbf{k}_\parallel=0} = \left.\frac{\partial^2 k_i}{\partial k_y^2}\right|_{\mathbf{k}_\parallel=0} > 0. \tag{8}$$

We now evaluate the total conductances by assuming that $C_{AP}(\mathbf{k}_\parallel)$ can be approximated by some constant value. In fact it does not matter what value we assume, as this term vanishes from the asymptotic expression for the TMR. This gives

$$\begin{aligned}\mathcal{G}_P &= e^{-2k_1^0 N}\int d\mathbf{k}_\parallel (A_0 + A_2\mathbf{k}_\parallel^2)\exp(-H_1\mathbf{k}_\parallel^2) \\ &= \pi e^{-2k_1^0 N}\left(\frac{A_0}{H_1 N} + \frac{A_2}{H_1^2 N^2}\right)\end{aligned} \tag{9}$$

for the parallel configuration and

$$\begin{aligned}\mathcal{G}_{AP} &= C_{AP}e^{-2k_2^0 N}\int d\mathbf{k}_\parallel \exp(-H_2\mathbf{k}_\parallel^2 N) \\ &\quad + e^{-2k_1^0 N}\int d\mathbf{k}_\parallel [B_2\mathbf{k}_\parallel^2 + B_4(k_x^4 + k_y^4)]\exp(-H_1\mathbf{k}_\parallel^2 N) \\ &= \frac{C_{AP}\pi e^{-2k_2^0 N}}{H_2 N} + \pi e^{-2k_1^0 N}\left(\frac{B_2}{N^2 H_1^2} + \frac{3B_4}{2N^3 H_1^3}\right)\end{aligned} \tag{10}$$

TABLE I. Asymptotic TMR coefficients for Fe/MgO/Fe.

	A_0	A_2	B_2	B_4	H_1
Fe/MgO/Fe	0.21404	2.71788	3.32082	67.8416	11.674

for the antiparallel configuration. These two equations show that $\mathcal{G}_{AP}$ decays faster in N than $\mathcal{G}_P$.

We now calculate the so-called "optimistic" TMR defined in Eq. (1), and perform an asymptotic expansion in powers of $1/N$. The term coupling to C_{AP} vanishes in this analysis since it decays with a larger negative exponent than the other terms. The result is

$$\mathrm{TMR} = \frac{A_0 H_1}{B_2}N - \frac{1}{2B_2^2}(2B_2^2 - 2A_2B_2 + 3A_0B_4) + O(1/N). \tag{11}$$

We observe that the the asymptotic expansion of the TMR is linear in MgO thickness and that the linear and constant coefficients depend only on terms coupling to the smallest MgO Fermi sheet k_1.

We now compare the analytic formula for the TMR with a fully realistic, numerical, tight-binding computation for the Fe/MgO/Fe tunneling junction. Details of the numerical computation can be found in Refs. 1 and 7. The amplitudes $A(\mathbf{k}_\parallel)$ and $B(\mathbf{k}_\parallel)$, depicted in Fig. 4, are obtained by fitting the expressions (3) and (5) to the conductances depicted in Fig. 3. [We do this for $N=15$ atomic planes of MgO so that the second term in Eq. (5) is negligible.] The constants A_0, A_2, B_2, and B_4 are obtained by fitting these amplitudes to the expressions (4) and (6). These values, along with $H_1 = \frac{\partial^2 k_1}{\partial k_x^2}|_{\mathbf{k}_\parallel=0}$, obtained from the complex Fermi surface of MgO, are given in Table I.

Finally, in Fig. 5 we compare the asymptotic formula with the full numerical calculation. The asymptotic formula shows fair agreement from about ten atomic planes of MgO and excellent agreement after about 15 atomic planes.

It can be seen from Eq. (11) that the TMR can only saturate in MgO thickness if the conductance in the parallel con-

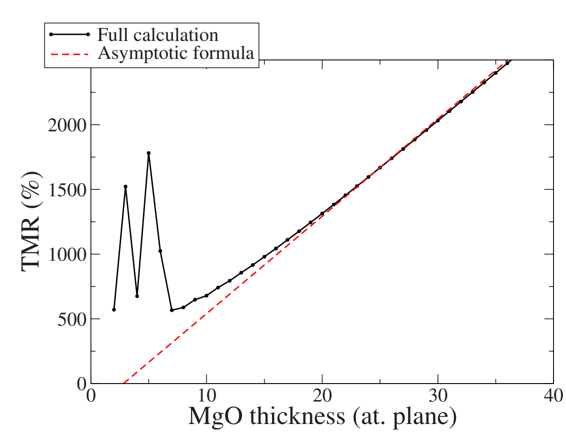

FIG. 5. (Color online) Comparison of the full numerical calculation for the TMR and the asymptotic formula given by Eq. (11) for Fe/MgO/Fe.

figuration were to vanish at the $\bar{\Gamma}$ point, i.e., $A_0=0$. Note also that the gradient of the TMR is inversely proportional to the curvature of the antiparallel conductance through the k_1 band at $\mathbf{k}_{\parallel}=0$ (B_2). This is to be expected, since the smaller B_2, the larger the region around the $\bar{\Gamma}$ point where conductance through k_1 is forbidden, and hence the larger the expected TMR.

We now briefly consider the effect of interfacial roughness. It follows from the discussion in Ref. 7 that the principle effect of interfacial roughness is to scatter electrons over the whole 2D Brillouin zone. This mechanism opens up the k_1 (Δ_1) conductance channel at the $\bar{\Gamma}$ point in the antiparallel configuration. This can be simulated in our calculation by adding a constant term B_0 to Eq. (6), which is related to the scattering strength to (and away from) the $\bar{\Gamma}$ point. If we include this extra term, it can be shown that the TMR has the form $(A_0-B_0)/B_0+O(1/N)$, and hence it saturates.

In conclusion, we have shown that for an epitaxial tunneling junction based on an MgO barrier, the optimistic TMR eventually grows linearly with the barrier thickness. This linear dependence is in complete agreement with previously published fully numerical calculations.[7,8] We have derived a simple asymptotic formula for the TMR as a function of barrier thickness and shown that this gives excellent agreement with full numerical calculations, performed here on Fe/MgO/Fe. As well as providing a definitive answer to the question of how the TMR grows with increasing MgO thickness, it is clear that Eq. (11) provides a valuable test to check that numerical studies are converging to the correct answer. The method we have used should be applicable to any epitaxial tunelling junction where each $\mathbf{k}_{\parallel}$ channel is independent. The result of TMR growing linearly with barrier thickness will apply to any epitaxial barrier with perfect spin filtering at the $\bar{\Gamma}$ point, e.g., Co/MgO/Co.

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[11] It has been suggested (Ref. 2) that these real sheets are responsible for the oscillations in TMR sometimes observed in experiment (Refs. 3, 9, and 10). However, as many authors have commented, since the real sheets do not occur near the $\bar{\Gamma}$ point, the oscillations associated with them must decay very quickly and so cannot be those observed. The origin of the oscillations seen in experiment remains a mystery, and is not discussed further in this communication.