Origin of the breakdown of Wentzel-Kramers-Brillouin-based tunneling models

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(Received 26 September 2006; published 15 December 2006)

The tunneling conductance of three varieties of CoFeB/MgO/CoFeB magnetic tunnel junctions depends quadratically on the applied voltage to anomalously high biases. Within the framework traditional of WKB models, this implies unphysical tunnel barrier parameters: heights near 20 eV, or widths corresponding to fewer than two MgO lattice constants. We demonstrate that the failure of such models to yield physically reasonable parameters originates from an experimentally unavoidable distribution of barrier thicknesses, possibly acting synergistically with the band structure of the barrier material. This implies that existing WKB models may lead to physically incorrect barrier parameters for contemporary tunnel junctions, magnetic or otherwise.

DOI: 10.1103/PhysRevB.74.212404 PACS number(s): 73.40.Gk, 72.25.Mk, 85.75.−d

Contemporary advances in spin-polarized electron tunneling have been realized with magnetic tunnel junctions (MTJs) that incorporate crystalline magnesium oxide as the barrier material.[1,2] The parameters of the tunneling barrier are interesting for fundamental physics, and are important for developing spintronics technologies, such as magnetoresistive random access memory (MRAM).[3] Experimental tunneling characteristics are generally fit to WKB-based models, such as those of Brinkman, Dynes, and Rowell (BDR),[4] and Simmons,[5] using the thickness (s) and interfacial barrier heights (ϕ_1 and ϕ_2) as adjustable parameters. These two models have nearly 1500 combined citations to date, and have been employed in normal metal, superconducting, semiconducting, molecular, and magnetic tunnel junctions composed of literally hundreds of different electrode and barrier material combinations. This work shows that parameters extracted from conventional fits with these omnipresent WKB models are physically unreasonable, and that this problem is resolved by incorporating reasonable assumptions about the nature of the barrier.

The tunneling conductance of three types of CoFeB/MgO/CoFeB devices shows a quadratic bias dependence that persists to anomalously high biases, where it should begin to increase exponentially within the framework of existing theories. The ubiquitous *quadratic expansion* of the BDR model results in statistically good fits, but strong disagreements arise when parameters obtained in this fashion are directly inserted into the full BDR model. This inconsistency implies that commonly used procedures based on traditional WKB models are inappropriate for contemporary tunnel junctions. We demonstrate that the origin of this behavior is a realistic and unavoidable amount of interfacial roughness, possibly in conjunction with the tunneling electron sensing the band structure of the barrier material. The latter may underscore the imminent necessity of first-principles analyses of modern tunneling devices, which has profound implications for barriers as simple as MgO and as complex as molecules.

Magnetic tunnel junctions composed of pinned CoFe/Ru/CoFeB synthetic antiferromagnets (SAF), MgO tunneling barriers, and free CoFeB/Ru/CoFeB SAFs were grown by magnetron sputtering at 0.1–0.5 Å/s.[6] The tunneling barrier was formed by *in situ* plasma oxidation of 16 Å metallic Mg films, which should yield approximately a 13 Å MgO barrier with lattice constant $a=4.2$ Å and a 3.7 eV barrier height for ideal growth.[7] The growth and annealing of these devices produces (001) textured MgO barriers.[1,8] The tunneling conductance ($G \equiv dI/dV$) was obtained numerically from room temperature I-V measurements, and independently verified by dV/dI measurements with a high-resolution resistance bridge using standard lock-in techniques; the latter was used to measure the temperature dependence down to 5 K. The dc bias was applied to the free layer with the pinned layer grounded, and all measurements were performed in remanence. The zero-bias resistance-area products in the parallel and antiparallel magnetic states were 2.3 and 5.1 kΩ μm^2 at 300 K, resulting in a tunneling magnetoresistance of 120%. All devices satisfied the MTJ tunneling criteria,[9] proving that tunneling is the dominant conduction mechanism.

Figure 1 shows that the experimental parallel state conductance (G_p) depends quadratically on the applied voltage in the entire bias range studied. It is commonplace to fit parabolic conductance data with an expansion to quadratic order in bias of the BDR model to determine the barrier parameters. This procedure results in a good fit ($\chi^2=1.29$) that yields $s=7.3$ Å, $\phi_1=2.2$ eV, and $\phi_2=2.9$ eV. The best-fit thickness is about 60% of the expected 13 Å, and the heights are within the expected range for real MgO barriers. However, when these best-fit results are used as input parameters for the full BDR model, a severe deviation from the data is found for biases above ~0.5 V (black line). This

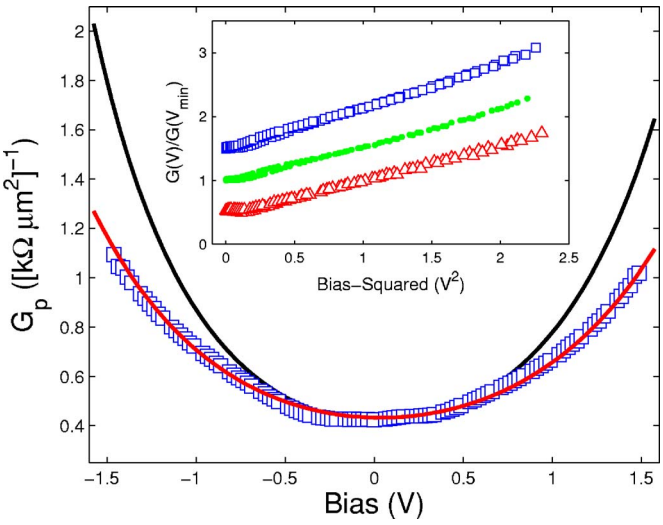

FIG. 1. (Color online) Experimental conductance (squares), best-fit using the full BDR model (red/grey line), and the full model calculation using the best-fit results via the quadratic expansion as input parameters to Eq. (1) (black line). The lowest order polynomial that reproduces the full model fit in this bias range is of sixth-order. Inset: Normalized conductance vs bias squared for devices with three different oxidation or anneal recipes (shifted for clarity) shows parabolic dependence over a large high bias range. From top to bottom, the data are from wafers B, A, and C (see Table 1).

represents a trend toward the exponential bias dependence that characterizes the Fowler-Nordheim tunneling regime (i.e., field emission). The quadratic expansion using these parameters is thus invalid above ~0.5 V, despite the high quality fit.

These observations are not isolated to a few samples. The inset of Fig. 1 shows the bias dependence (including both polarities) of the normalized conductance for three sets of 26 junctions each, which differ only in their oxidation and anneal procedures. The bias dependence was independent of junction shape, area, and absolute resistance. The bias axis has been shifted by the conductance minimum to emphasize the intrinsic symmetry of the conductance.[10] The linearity shows that the conductance was approximately quadratic for most of the bias range. All three device types exhibited low bias deviations from quadratic behavior in the range 100–150 meV, which have been attributed to magnon excitations in the ferromagnetic electrodes.[11] As with the above example, fitting these data with the quadratic expansion yields statistically good fits with reasonable barrier heights and thinner barriers than expected from the growth. In all cases, when these parameters are inserted into the full model, a severe discrepancy is found that is similar to that shown in Fig. 1.

Following BDR, the tunneling current density in the WKB approximation for asymmetric barriers is given by

$$j = \frac{2e}{h}\int\int \exp\left(-\frac{2}{\hbar}\int_0^s \sqrt{2m\phi(x,V,E)}dx\right) \times[f(E(\vec{k})) - f(E(\vec{k}) - |e|V)]dEd\vec{k}. \quad (1)$$

Here, s is a single barrier thickness, $\phi(x,V,E)$ is the position-dependent barrier height at bias V for an electron with incident energy E, e is the elementary charge, m is the free-electron mass, $\vec{k}$ are the wave vectors parallel to the junction interface, and f is the Fermi function. Because of its simplicity and robust generality, an expansion of the BDR model to quadratic order in bias [Eq. (7) in Ref. 4] has become ubiquitously used to extract tunnel barrier parameters. However, by the very nature of its being an expansion, *this is valid over a very limited bias range, typically less than one-third of the barrier height*. An analysis of this model reveals that the intrinsic barrier height must be nearly 20 eV to sustain parabolic conductance up to 1.5 V with a 15 Å barrier. This is clearly unphysical since the band gap of MgO is only about 7.4 eV,[7] the expected barrier height is half this, and the work functions of most metals are below 6 eV. If we conversely suppose an ideal 3.7 eV barrier, a 5 Å barrier is necessary to have parabolic dependence to 1.5 V. Even if a barrier this thin (1.2 MgO lattice constants) could be produced reliably without pinholes over a realistic area, dielectric breakdown would limit such devices to biases less than a volt.[12] We are thus led to the distinct conclusion that the full model must be used to analyze data when the bias range is on the order of the barrier height.

Several possible causes for these discrepancies can be easily eliminated. Thermal smearing cannot cause this behavior because it should affect the conductance only within a fraction of a volt (a few kT).[13] Thermal smearing decreases with decreasing temperature, but measurements down to 5 K show no significant temperature dependence. Likewise, effects due to the image potential[4] are unlikely since this reduces both the barrier width and height, which would cause deviations from parabolic dependence at even *lower* applied biases. Neither a series resistance associated with the leads nor heating were significant because this behavior was independent of geometry and absolute junction resistance. We cannot, however, rule out an unexpected intrinsic interfacial series resistance between the CoFeB and MgO. Such a resistance would go undetected with standard scaling arguments, but would result in a stretched bias scale if a significant voltage drop occurred at one or both interfaces. The total interfacial series resistance required for such an artifact is about one-third of the junction resistance, ~0.5 kΩ μm^2 per interface. This region would have a resistivity around 10^8 $\mu\Omega$ cm if it were 10 Å thick and Ohmic. Segregation of boron or its oxides at the interfaces could be the cause,[14,15] though this extended parabolic dependence has been observed, perhaps unbeknownst to the authors, in devices without boron (see e.g, Refs. 16–18). Moreover, such a thin boron rich area could become part of the tunneling barrier, which, contrary to the best-fit results, would result in a thicker barrier.

Two realistic possibilities that cannot be dismissed are the influence of the band structure of the barrier material, and an unavoidable roughening of the heterostructure interfaces. The importance of the band structure of crystalline tunnel barriers, MgO in particular, has been recently emphasized.[19,20] The MgO band structure can be addressed in an unconventional way by parametrizing the BDR model with the electronic effective mass. While it is known that m^*/m_e is important in the Fowler-Nordheim regime because

TABLE I. Best-fit barrier parameters using the full BDR model for three device types. Bold text indicates parameters that were fixed during the fits.

Wafer	s (Å)	ϕ_1 (eV)	ϕ_2 (eV)	m^*/m_e	χ^2
A	6.25	2.96	3.78	**1.0**	1.14
B	6.59	2.89	3.65	**1.0**	1.64
C	6.33	3.66	3.47	**1.0**	1.61
A	8.52	2.33	3.00	**0.4**	1.14
B	8.93	2.28	2.95	**0.4**	2.54
C	8.64	2.89	2.74	**0.4**	1.67
A	**13**	1.66	2.18	0.11	1.71
B	**13**	1.72	2.19	0.13	3.25
C	**13**	2.03	2.06	0.12	2.18

electrons tunnel into the conduction band of the barrier,[21] we are unaware of any studies showing its importance in the pure tunneling regime.

Three different full BDR model fit procedures were used to investigate the effect of m^*/m_e on the conductance (Table I). The three procedures differed only in the parameters that were fixed and free during the fits, and were performed on the data sets of Fig. 1. First, a traditional fit was performed using m^*/m_e fixed at 1, while s, ϕ_1, and ϕ_2 were free parameters. This yields a best fit s of ~1.5 MgO lattice constants (~6 Å), and near ideal ϕ for one of the heights for each device type. This s is very different from what is expected from preparation (calibrated growth rates and oxide formation). Moreover, all devices resist dielectric breakdown to nearly 2 V at room temperature, which earlier experimental results imply is quite unexpected for such a thin barrier.[6,12] As for the near ideal ϕ, a recent study by Mather *et al.* showed that comparable MgO barriers had band gaps that were significantly lower than that of bulk MgO, implying that ideal behavior is highly unlikely for thin tunnel barriers.[22] Based on these arguments, we believe that the best-fit parameters extracted from traditional methods using the existing full WKB models are physically incorrect.

For the second procedure, m^*/m_e was fixed at the expected value of 0.4, with all other parameters free. Relative to the traditional fit, this yields a thicker barrier of 2.1 MgO lattice constants (~9 Å), with lower than ideal heights (2.3–3.0 eV). This best-fit barrier thickness is still thinner than expected, and the heights are larger than those found by Mather *et al.* for similar barriers. In the final procedure, the known thickness was taken as a fixed parameter, while m^*/m_e, ϕ_1, and ϕ_2 were free. In this case, the best-fit barrier heights are reasonable, but the best $m^*/m_e \approx 0.12$ is unexpectedly small. It is worth noting that much worse quality fits ($\chi^2 > 1000$) resulted when adjusting only the heights with s and m^*/m_e simultaneously fixed at 13 Å and either 1 or 0.4, respectively.

Based on the resulting χ^2 values from these three procedures, the best fits for all device types are found using a fixed free mass. However, the χ^2 with $m^*/m_e=0.4$ are very similar to those with the free mass for two MTJ types. This similarity makes it difficult to determine whether it is best to use 1 or 0.4 for m^*/m_e. The latter is potentially more appealing because of the thicker s and lower ϕ. This implies that the tunneling electron may sense not only the magnitude of the insulator's band gap, but also details of its overall band structure.

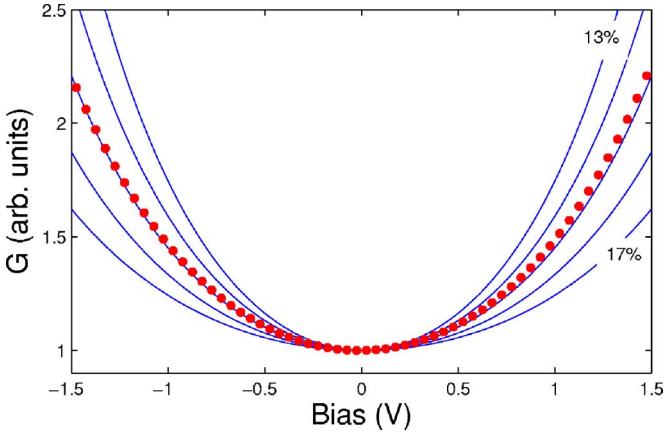

FIG. 2. (Color online) Comparison of the best-fit single thickness model for wafer C with $m^*/m_e=1$ (red points) and the full model calculation assuming $\bar{s}=13$ Å and roughness parameters of 13–17% of $\bar{s}$ in 1% steps (top to bottom).

The failure of the model to recover the expected thickness can be attributed to interfacial roughness, which, contrary to popular belief, cannot be avoided even in samples grown by molecular beam epitaxy.[23] To simulate the effect of roughness, we calculated the current density via Eq. (1) for a distribution of barrier thicknesses s_n with symmetric barrier heights, weighted each individual current density $j(s_n)$ depending on its thickness, and summed these as parallel conduction channels. The net conductance G_{net} was obtained by differentiating the net current density $j_{net}=\Sigma_n \alpha(s_n) j(s_n, \phi, V)$, with the nth conduction channel weighted by a coefficient $\alpha(s_n)$ that is obtained from a Gaussian distribution of thicknesses centered at mean thickness $\bar{s}$ with the standard deviation σ defining the roughness. The distribution $s_n=\bar{s}\pm\sigma$ represents variations of the barrier thickness over the entire junction area, and at both interfaces.

Using the data of wafer C with a nearly symmetric barrier as an example, Fig. 2 shows that these data are well fit ($\chi^2=1.14$) using $m^*/m_e=1$, $\phi=3.5$ eV, and a distribution 13 Å±15%. This is about 2 Å of roughness, which is less than half an MgO lattice constant. A similar analysis of these data using the best-fit full model calculation with $m^*/m_e=0.4$ yields a thickness distribution 13 Å±17%. In both cases, the required amounts of roughness are experimentally reasonable, and are predicted by sophisticated growth models.[24] Note that the best-fit thickness with the full BDR model assuming a single thickness is $2-3\sigma$ below the mean thickness. This together with the limited range of roughness presented in Fig. 2, demonstrates that interfacial roughness cannot be neglected because of the extreme sensitivity of the net conductance to this undeniable parameter.

In summary, the tunneling conductance of magnetic tunnel junctions was found to be parabolic to anomalously high

biases in a large variety of tunnel junctions with MgO barriers. The data are fit well by the quadratic expansion of the BDR model throughout the entire bias range, but the parameters obtained this way produce serious discrepancies when inserted into the full functional form of the model. This inconsistency proves that expansions of traditional tunneling models are not valid in the high bias ranges that are achieved by today's state-of-the-art devices. Within this framework, accounting for the barrier band structure by parametrizing the model with the effective mass provides equally good fits, but arguably more realistic barrier parameters relative to the traditional method that assumes the free-electron mass. This indicates that the tunneling electron may be sensitive to the band structure of the barrier, which may suggest the imminent necessity for first-principles analyses of contemporary tunneling systems. Despite the goodness of these fits, existing theoretical models fail to yield rational barrier parameters, particularly for the thickness, even when the band structure of the insulator is considered. The origin of this collective failure was traced to the assumption of a single barrier thickness, perhaps working in collaboration with the band structure of the barrier. Introducing a distribution of barrier thicknesses consistent with an experimentally feasible amount of interfacial roughness allows the prepared barrier thickness to be recovered, and stresses the importance of this unavoidable characteristic of real tunneling junctions.

We acknowledge support by the US Department of Energy (UCSD), the Swedish Foundation for Strategic Research, The Swedish Research Council, and The Göran Gustafsson Foundation (J.Å.). The authors thank R. C. Dynes, J. M. Rowell, N. Newman, A. J. Leggett, W. H. Butler, M. Chshiev, R. Escudero, and H. Dery for stimulating discussions.

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