Large magnetoresistance in Fe/MgO/FeCo(001) epitaxial tunnel junctions on GaAs(001)

M. Bowen,[a] V. Cros, F. Petroff, and A. Fert
Unité Mixte de Physique CNRS-THALES, Domaine de Corbeville, 91404 Orsay Cedex, France and Université Paris Sud, 91405 Orsay Cedex, France

C. Martínez Boubeta, J. L. Costa-Krämer, J. V. Anguita, A. Cebollada, and F. Briones
Instituto de Microelectrónica de Madrid-IMM (CNM-CSIC), Isaac Newton 8-PTM, 28760 Tres Cantos, Madrid, Spain

J. M. de Teresa, L. Morellón, and M. R. Ibarra
Instituto de Ciencia de Materiales de Aragón, Universidad de Zaragoza-CSIC, Facultad de Ciencias, 50009 Zaragoza, Spain

F. Güell, F. Peiró, and A. Cornet
Electronic Materials and Engineering, Facultat de Fisica, Universitat de Barcelona Av. Diagonal 645-647, E-08028 Barcelona, Spain

(Received 19 February 2001; accepted for publication 30 July 2001)

We present tunneling experiments on Fe(001)/MgO(20 Å)/FeCo(001) single-crystal epitaxial junctions of high quality grown by sputtering and laser ablation. Tunnel magnetoresistance measurements give 60% at 30 K, to be compared with 13% obtained recently on (001)-oriented Fe/amorphous-Al_2O_3/FeCo tunnel junctions. This difference demonstrates that the spin polarization of tunneling electrons is not directly related to the density of states of the free metal surface—Fe(001) in this case—but depends on the actual electronic structure of the entire electrode/barrier system. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1404125]

The magnetoresistance of magnetic tunnel junctions (MTJs)[1] is of uncontested interest for important applications with, in particular, promising perspectives for the fabrication of nonvolatile memories (Magnetic RAM).[2] Up to now, most studies have been performed on MTJs with a layer of amorphous alumina as insulating barrier between the ferromagnetic electrodes, yielding large and reproducible tunneling magnetoresistance (TMR). However, from a fundamental point of view, i.e., the understanding of the physics of spin-dependent tunneling, a transport study through an amorphous insulator is hardly accessible in a theoretical approach. Most numerical calculations of spin-dependent tunneling and TMR have been developed for single crystal MTJs such as $Co/Al_2O_3/Co(100)$,[3] Fe/ZnSe(100),[4] or Fe/MgO(100).[5,6] All emphasize that a correct depiction of the spin-dependent tunneling properties of epitaxial MTJs must transcend the simple potential barrier image and take into account the interplay of electronic structure between metal and insulator. A test of these models can be performed on single-crystal epitaxially grown structures. Towards this end, much work has been expended to characterize the growth and electrical behavior of ultrathin MgO layers.[7] In this letter, we present experimental results showing large TMR values in Fe(001)/MgO(001)/FeCo(001) epitaxial tunnel junctions.

FeCo/MgO/Fe epitaxial structures were grown on GaAs(001) in a combined sputtering/laser ablation system with a base pressure of 2×10^{-9} mbar as described elsewhere.[8] We use a MgO buffer layer as an interdiffusion barrier with good electrical insulation characteristics.[9] This prevents the incorporation of As from the substrate into the Fe bottom electrode—an important precaution since, in a subsequent step, the MgO barrier is grown at 400 °C in order to obtain good crystallinity. This MgO buffer layer also offers an appropriate symmetry and lattice match for the epitaxy of Fe on top, with the well known orientation relation Fe(100)[001]//MgO(100)[110]. $Fe_{50}Co_{50}$ and Fe layers were deposited from individual Fe and Co targets by triode sputtering with an Ar pressure of 4×10^{-4} mbar and with deposition rates in the range of 0.1–0.3 Å/s. Optimal deposition temperatures were 400 °C for the MgO, RT for FeCo top electrode, and RT plus annealing at 400 °C for the Fe bottom electrode. This low-temperature deposition and subsequent annealing process leads to an optimal Fe electrode in terms of crystallinity, continuity, and interface sharpness.

Figure 1(a) shows RHEED patterns for a typical $Fe_{50}Co_{50}$/MgO/Fe structure. $Fe_{50}Co_{50}$ and Fe layers are bcc structured with sharp diffraction lines for both azimuths. Similar information about epitaxial quality could be concluded from the MgO barrier pattern. Further x-ray diffraction symmetric and asymmetric scans confirm that the whole structure is epitaxial with lattice parameters close to bulk values. To check the continuity of the layers and the sharpness of the interfaces, a specific multilayered Fe/MgO test structure was grown in conditions described above with nominally constant 80-Å-thick Fe metallic layers separated by increasingly thin MgO layers, from 80 to 20 Å. Figure 1(b) presents a [110] cross section transmission electron microscopy (TEM) image from this test structure covering a lateral region of about 1 μm—dark regions correspond to the Fe layers while the lighter regions reflect the MgO layers. Continuous and good crystal quality MgO films with sharp

[a]Electronic mail: martin.bowen@thalesgroup.com

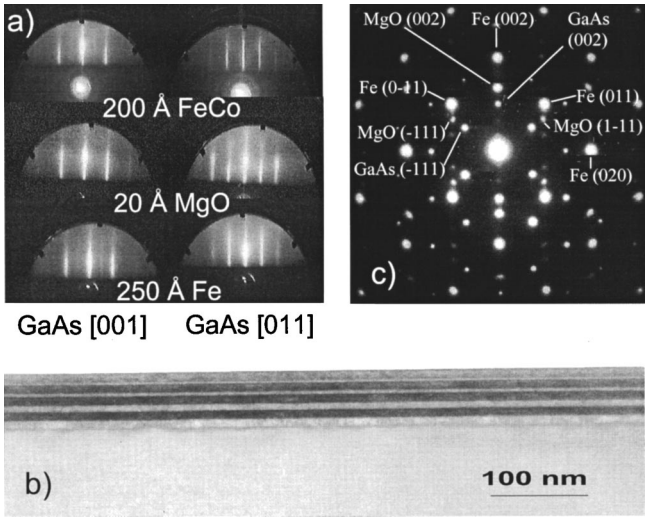

FIG. 1. (a) RHEED patterns along the GaAs [100] and [110] substrate azimuths of a typical FeCo/MgO/Fe epitaxial structure; (b) cross section TEM image of a Fe/MgO(001) multilayer with gradually decreasing MgO layer thickness; (c) TEM electron diffraction pattern.

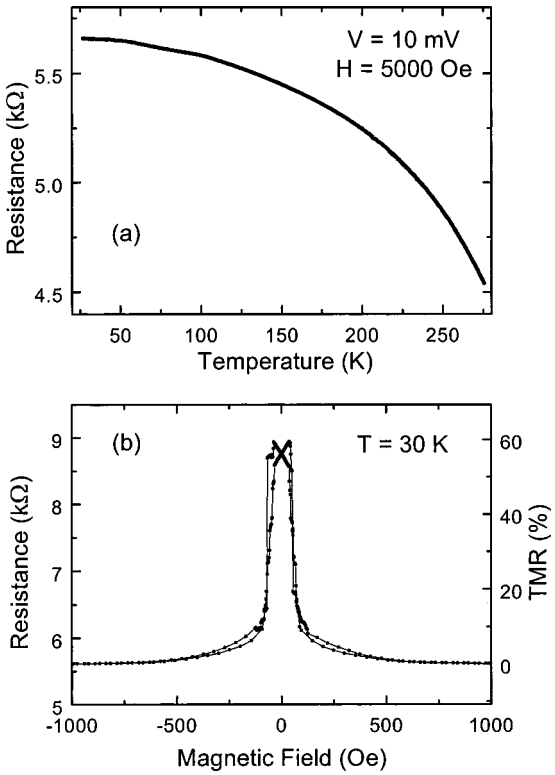

FIG. 2. Transport studies for a Fe(001)/MgO 20 Å(001)/FeCo(001) tunnel junction of diameter 10 μm: (a) resistance vs temperature at $V=10$ mV and $H_{app}=5000$ Oe; (b) resistance and TMR vs magnetic field at $V=10$ mV and $T=30$ K.

interfaces are obtained all the way down to the thinnest oxide layer. The electron diffraction pattern for a selected area of the TEM image shown in Fig. 1(b) illustrates the aforementioned orientation relationship Fe(100)[001]// MgO(100)[110] and is indicative of the crystallinity and high quality of the structure.

A Fe(200 Å)/MgO(20 Å)/FeCo(250 Å) trilayer grown epitaxially onto MgO(001)-buffered GaAs(001) was processed by optical lithography.[10] Here we present results obtained on a tunnel junction of diameter 10 μm. Transport measurements were performed in four-point voltage source mode ($V^+=$Fe). The resistance of the Fe electrode is more than 100 times smaller than the junction resistance, thus ruling out any significant contribution from geometrical effects.[11] As shown in Fig. 2(a), the resistance of our MTJ (at 10 mV) saturates below 50 K and then decreases slowly by about 25% between 50 and 300 K. This is a typical temperature dependence in which intrinsic tunneling transport processes have given way above 50 K to additional thermally assisted processes. The temperature dependence of $I(V)$ curves places[12] the barrier height at $\phi=0.9$ eV. Simmons' equations[13] yield $\phi\sim1.1$ eV for a barrier thickness $d\sim15$ Å, in good agreement with previous transport studies using epitaxial MgO(111) by Kiyomura *et al.*,[14] and polycrystalline MgO by Moodera *et al.*,[15] who both report $\phi\sim0.9$ eV using this method. This value is lower than half of the 5.5 eV MgO band gap calculated for an ultrathin layer.[5] This difference may be due to metal-induced gap states in the MgO barrier[4] although we cannot completely rule out the presence of stoichiometric and/or thickness inhomogeneities in the insulating film. A somewhat higher value of ϕ has been reported by Wulfhekel *et al.* through STM measurements in which both the MgO and the vacuum barriers are taken into account.[7]

Figure 2(b) shows an $R(H)$ cycle taken at 30 K for an applied bias of +10 mV. We find a TMR of +60%, using the definition TMR$=(R_{AP}-R_P)/R_P$. The rise in resistance to the antiparallel (AP) state in a decreasing field before reaching $H=0$, and more generally the symmetry of the $R(H)$ curve around $H=0$, is the result of overmilling into the bottom Fe electrode during the junction mesa definition, thus creating a stray field-induced antiparallel state. As temperature increases, the TMR decreases in almost linear fashion to 27% at 300 K. The low-temperature TMR value is in agreement with expectations from Jullière's expression[16] if $P_{Fe}=45\%$ and $P_{FeCo}=51\%$, which are the highest values of P found in recent experiments, are used.[17] It is worth noting that these polarization values were obtained in tunnel junctions with polycrystalline electrodes and an amorphous barrier. Yuasa *et al.* have studied Fe/Al_2O_3/FeCo tunnel junctions[18] with (100)-, (110)-, and (211)-oriented single-crystal electrodes and amorphous Al_2O_3 yielding up to 40% TMR for the (211) orientation but only 13% for the (100) orientation. Yuasa *et al.* ascribed this weak TMR to a small (7%) spin polarization of the calculated density of states (DOS) at the (100) surface of Fe. The 60% TMR we find for Fe(100) demonstrates that the spin polarization of tunneling electrons cannot be directly correlated with the spin-polarized DOS of a free metal surface, but depends on the actual electronic structure of the barrier/electrode system and can be quite different for Fe(100)/Al_2O_3 and Fe(100)/MgO(100) interfaces.

In this vein, recent spin-polarized tunneling experiments[19] have emphasized the role of the metal–oxide interface in favoring a particular spin polarization and electronic character of the tunneling current. Co/Al_2O_3 interfaces result in a positive polarization which, in an oversimplified picture, can be ascribed to a predominant tunneling of *s*-character electrons due to a specific bonding mechanism at the interface.[3] On the other hand, a Co/$SrTiO_3$ interface leads

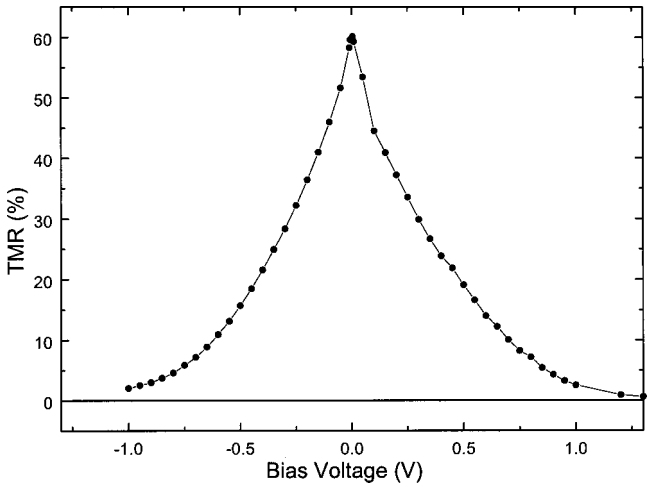

FIG. 3. Bias voltage dependence of the TMR at $T=30$ K for a Fe(001)/MgO 20 Å(001)/FeCo(001) tunnel junction of diameter 10 μm. TMR values were obtained from $R(H)$ data. $V^+=$Fe.

to a negative polarization corresponding to the sign of the polarization at E_F in the *d* band of Co. The complex bias dependence of the TMR with a $SrTiO_3$ barrier also differs from the rapid and symmetric decrease observed in any junction with an Al_2O_3 barrier. We present a bias dependence study of the TMR obtained for our Fe/MgO/FeCo junction in Fig. 3. The TMR decreases almost symmetrically from a value of 60% at 10 mV to nearly 0 around 1.4 V. This result is confirmed with 2 mV resolved $I(V)$ curves taken in the parallel and antiparallel states. Given the similarity of the symmetric decrease of the TMR with bias for MgO and Al_2O_3, we surmise that *s*-character electron transmission occurs predominantly for a MgO barrier. The lack of *d* elements in MgO and Al_2O_3 lends credence to this hypothesis. As corroborating evidence, Butler *et al.* point out in recently published first principles conductance calculations[5] for Fe(100)/MgO(100)/Fe(100) trilayers that, despite a strongly negative spin polarization on the interfacial Fe layer, the spin polarization of tunneling electrons should be positive due to wave function symmetry matching in the Fe electrodes and in the MgO barrier. Tunneling conductance in the parallel alignment of Fe(100) electrodes is dominated by the majority spin channel owing to a state (Δ_1) of significant *s* character which decays much more slowly than other states and does not exist in the minority spin channel. This state will further accentuate the tunneling spin polarization in the parallel state, and the resulting TMR, as barrier thickness is increased—a discrepancy to Jullière's time-proven model which is only governed by the spin polarization of the tunnel junction electrodes. Numerically, the TMR values calculated by Butler *et al.* are much higher (2000% for 20 Å MgO) than our experimental value of 60% (with a FeCo top electrode), as is generally the case when comparing theory with experiment. More interestingly, in future experiments we will aim to confirm the predicted relative increase of the TMR with MgO barrier thickness.

In conclusion, we have observed tunneling MR on Fe(001)/MgO(20 Å/FeCo(001) grown by a combination of laser ablation and triode sputtering onto MgO-buffered GaAs(001). As evidenced by RHEED, x-ray diffraction and TEM analyses, optimized growth conditions result in entirely epitaxial samples of high crystalline quality with flat, sharp interfaces. Transport measurements show 27% TMR at 300 K which increases to 60% at 30 K. We construe from the bias dependence of the TMR, previous experimental results and recent calculations[5] that *s*-character electrons are predominantly tunneling in the case of a 20 Å MgO barrier.

The Spanish Commission of Science and Technology (CICyT), Comunidad de Madrid and Acciones Integradas Hispano Francesas (Picasso Program) are acknowledged for financial support.

[1] J. S. Moodera, L. R. Kinder, T. M. Wong, and R. Meservey, Phys. Rev. Lett. **74**, 3273 (1995).
[2] S. S. P. Parkin, K. P. Roche, M. G. Samant, P. M. Rice, R. B. Beyers, R. E. Scheuerlein, E. J. O'Sullivan, S. L. Brown, J. Bucchigano, D. W. Abraham, Y. Lu, M. Rooks, P. L. Trouilloud, R. A. Wanner, and W. J. Gallagher, J. Appl. Phys. **85**, 5828 (1999).
[3] I. I. Oleinik, E. Yu. Tsymbal, and D. G. Pettifor, Phys. Rev. B **62**, 3952 (2000).
[4] P. Mavropoulos, N. Papanikolaou, and P. H. Dederichs, Phys. Rev. Lett. **85**, 1088 (2000).
[5] W. H. Butler, X. G. Zhang, T. C. Schultess, and J. M. MacLaren, Phys. Rev. B **63**, 54416 (2001).
[6] J. Mathon and A. Umerski, Phys. Rev. B **63**, R220403 (2001); I. Mertig (private communication).
[7] W. Wulfhekel, M. Klaua, D. Ullmann, F. Zavaliche, J. Kirschner, R. Urban, T. Monchesky, and B. Heinrich, Appl. Phys. Lett. **78**, 509 (2001).
[8] C. Martínez Boubeta, E. Navarro, A. Cebollada, F. Briones, F. Peiró, and A. Cornet, J. Cryst. Growth **226**, 223 (2001).
[9] W. H. Lee, H. L. Cho, B. S. Cho, J. Y. Kim, W. J. Nam, Y.-S. Kim, W. G. Jung, H. Kwon, J. H. Lee, J. G. Lee, P. J. Reucroft, C. M. Lee, and E. G. Lee, Appl. Phys. Lett. **77**, 2192 (2000).
[10] F. Montaigne, J. Nassar, A. Vaurès, F. Nguyen Van Dau, F. Petroff, A. Schuhl, and A. Fert, Appl. Phys. Lett. **73**, 2829 (1998).
[11] R. J. M. Van de Verdonck, J. Nowak, R. Meservey, J. S. Moodera, and W. J. M. de Jonge, Appl. Phys. Lett. **71**, 2839 (1997).
[12] C. B. Duke, *Tunneling in Solids* (Academic, New York, 1969).
[13] J. G. Simmons, J. Appl. Phys. **34**, 2581 (1963).
[14] T. Kiyomura, Y. Maruo, and M. Gomi, J. Appl. Phys. **88**, 4768 (2000).
[15] J. S. Moodera and L. R. Kinder, J. Appl. Phys. **79**, 4724 (1996).
[16] M. Jullière, Phys. Lett. A **54**, 225 (1975).
[17] D. J. Monsma and S. S. P. Parkin, Appl. Phys. Lett. **77**, 720 (2000).
[18] S. Yuasa, T. Sato, E. Tamura, Y. Suzuki, and H. Yamamori, Europhys. Lett. **52**, 344 (2000).
[19] J. M. de Teresa, A. Barthélémy, A. Fert, J. P. Contour, F. Montaigne, and P. Seneor, Science **286**, 507 (1999).