Improved tunneling magnetoresistance at low temperature in manganese junctions grown by molecular beam epitaxy

R. Werner,1 A. Yu. Petrov,2 L. Alvarez Miño,3 R. Kleiner,1 D. Koelle,1 and B. A. Davidson2,a)
1Physikalisches Institut–Experimentalphysik II, Universität Tübingen, Auf der Morgenstelle 14, 72076 Tübingen, Germany
2TASC National Laboratory, CNR-IOM, S.S. 14 Km 163.5 in AREA Science Park, 34012 Basovizza, Trieste, Italy
3Universidad Nacional de Colombia, Sede Manizales, Cra 27 # 64-60 Manizales, Colombia

(Received 24 January 2011; accepted 31 March 2011; published online 20 April 2011)

We report resistance versus magnetic field measurements for a La0.65Sr0.35MnO3/SrTiO3/La0.65Sr0.35MnO3 tunnel junction grown by molecular-beam epitaxy, that show a large field window of extremely high tunneling magnetoresistance (TMR) at low temperature. Scanning the in-plane applied field orientation through 360°, the TMR shows fourfold symmetry, i.e., biaxial anisotropy, aligned with the crystalline axis but not the junction geometrical long axis. The TMR reaches ~1900% at 4 K, corresponding to an interfacial spin polarization of >95% assuming identical interfaces. These results show that uniaxial anisotropy is not necessary for large TMR, and lay the groundwork for future improvements in TMR in manganese junctions. © 2011 American Institute of Physics. [doi:10.1063/1.3581885]

The figure of merit for magnetic tunnel junctions (MTJs) is the tunneling magnetoresistance (TMR) ratio, which determines their performance in practical devices such as magnetic random access memories and low-field sensors.1 An MTJ consists of two ferromagnetic electrodes separated by a thin insulating tunneling barrier. According to the Julliere model,2 the TMR ratio is defined as $TMR = \frac{(R_{AP} - R_p)}{R_p} = 2P_1P_2/(1-P_1P_2)$. Here $P_1$ and $P_2$ are the spin polarizations of the two electrodes and $R_{AP}$ and $R_p$ are the junction resistances with antiparallel and parallel orientation, respectively. Accordingly, an MTJ made from half-metallic electrode materials, such as doped manganites,3 should yield an infinite TMR ratio at temperature $T$ well below the Curie temperature $T_C$. Noting that TMR is more precisely associated with the properties of the electrode/barrier interface,4 this concept has been extended to also describe interfaces as half-metallic,5 i.e., the TMR is determined by the spin-polarization of the local density of states at the two interfaces with the barrier. Ferromagnetic correlations at manganese surfaces and interfaces are known to be weaker than in bulk, causing a “dead layer.”6–8 For example, at the vacuum/La1-xSrxBaMnO3 (LSMO) interface the nonferromagnetic layer is about three unit cells (uc) thick at $T = 200$ K,9 well below the bulk $T_C \approx 360$ K for ferromagnetic LSMO (F-LSMO) with optimal doping $x = 0.35$. This and other effects have been discussed to explain the disappearance of the TMR well below the bulk $T_C$ in manganese MTJs.6,10–12 Attempts have been made to “engineer” the interfaces by creating a doping profile to overcome this problem, and even though the TMR ratio remained low,13 spectroscopic characterization suggested this approach could improve the low-temperature TMR.14

To date, TMR at small dc voltage bias of MTJs based on nonoxide electrodes reached ~1150% at $T = 5$ K (Ref. 15) while the highest ratio was reported for manganese/titanate interfaces with a maximum value of about 1800% at 4 K in a very small window of applied in-plane magnetic field $H$.16 Here, an antiferromagnetic CoO layer was used to pin the upper electrode via exchange bias17 that can favor uniaxial anisotropy in the pinned electrode; such anisotropy was claimed necessary for the stabilization of well-defined antiparallel states and high TMR ratios.11,16

In this letter, we report on the TMR of MTJs based on F-LSMO with an antiferromagnetic $x = 0.65$ LSMO (AF-LSMO) exchange bias layer and a SrTiO3 (STO) barrier, grown by molecular beam epitaxy (MBE). We find a TMR ratio up to ~1900% at $T = 4$ K, which decreases rapidly with increasing $T$, disappearing at ~280 K. Rotating the applied in-plane magnetic field, we find a fourfold symmetry of the TMR, indicating that uniaxial anisotropy is not required for high TMR ratios.

For sample fabrication, we developed atomic-layer control of LSMO and STO growth and their interfaces by combining reactive MBE (Ref. 18) with in situ reflection high-energy electron diffraction (RHEED) techniques, extending the work of Haeni et al.19 These RHEED techniques permit us to adjust the surface termination at any point during deposition, including during interface growth.20 LSMO/STO/LSMO trilayers were grown on (001)-oriented STO substrates at a typical substrate temperature $T_s = 750$ °C and ozone pressure $p = 10^{-6}$ mbar. The bottom and top F-LSMO electrode thicknesses were 50 uc, separated by a tunnel barrier of stoichiometric STO, 5–6 uc thick. A 100 uc thick AF-LSMO layer was grown underneath the bottom electrode to increase and shift its coercive field $H_c$ due to exchange bias. The resulting difference in $H_c$ between the electrodes favors the establishment of fully antiparallel magnetization orientation of the two electrodes in a larger window of $H$ (Ref. 17) in the resistance versus magnetic field $R(H)$ loops used to determine the TMR ratio. The effect of exchange bias on the junction TMR characteristics should only be seen below ~250 K, in agreement with exchange bias effects seen in the hysteresis loops of an AF-LSMO (100 uc)/F-LSMO (50 uc) junction.
uc) bilayer by superconducting quantum interference device magnetometry measured independently.

Vertical mesa MTJs were patterned in several steps by photolithography and Ar ion milling; for details see Ref. 21. Figure 1 shows in (a) an optical microscope image of an MTJ with a $5 \times 30 \mu m^2$ mesa with vertical current injection from a Au/Ti/Au top contact and the stacking sequence of the sample in (b).

The junction resistance was more than an order of magnitude larger than the electrode resistances, ensuring uniform injection of bias current $I$. Electrical transport measurements were made in 2- and 3-point geometries in a He-flow cryostat at $T=4–300$ K. A Helmholtz coil outside the cryostat allows full rotation of $H$ from $\alpha=0^\circ–360^\circ$ with a maximum amplitude $H_{\text{max}}=1$ kOe. The angle $\alpha$ describes the relative orientation of $H$ with respect to the long side of the junction (cf. Fig. 1). All $R(H)$ loops shown or discussed below were taken after the junction was field-cooled at $H_{\text{prech}}$ and the stacking sequence of the crystallographic lattice $a$-axis direction. The (differential) junction resistance was measured by a lock-in amplifier at a dc voltage bias $V=0$, using an ac current amplitude of a few nanoamperes. Full characterization of the junction $I(V)$ and TMR as a function of dc voltage bias will be reported separately.

The inset of Fig. 2 shows two representative $R(H)$ loops at low temperature ($T=4$ K) and $\alpha=145^\circ$, taken after two identical cooling cycles. We define $TMR(R)=[R(H=H_{\text{prech}})-R_{\text{min}}]/R_{\text{min}}$ and the maximum TMR ratio within an $R(H)$ loop as $TMR_{\text{max}}=(R_{\text{max}}-R_{\text{min}})/R_{\text{min}}$. Here, $R_{\text{max}}$ and $R_{\text{min}}$ are the maximum and minimum junction resistances, and only in the ideal case of uniform and fully antiparallel electrode magnetization does this definition coincide with TMR, defined above. The $R(H)$ loops differ in shape (asymmetry), $R_{\text{max}}$ and width $\Delta H$ of the magnetic field window, defined as the full width at half maximum of the $R(H)$ peaks. The $R(H)$ loop with larger $\Delta H$ shows a stronger asymmetry in peak heights and the highest $TMR_{\text{max}}=1904\%$, which also exceeds measured TMR ratios for other combinations of field orientations $\alpha_{\text{FC}}$ and $\alpha$, during cooling and measurement, respectively.

The $R(H)$ loops (cf. inset of Fig. 2) show switching to the high and low $R$ states at $\sim100$ Oe and 200–300 Oe, which we attribute to $H_c$ of the upper (free) and bottom (exchange-biased) electrodes, respectively. We note that these values of increased $H_c$ as compared to single layer F films and nearly negligible hysteresis loop shifts $H_{\text{prech}}$ for the bottom electrode due to an exchange bias field deduced from the inset of Fig. 2, are consistent with $H_{\text{prech}}$ and $H_c$ in exchange bias AF/F bilayers of the same thicknesses measured separately (not shown). The $R(H)$ loops for nominally identical cooling cycles implies that the exchange bias coupling at the AF/F interface can vary between cooling cycles. We attribute this to a history dependence of the exchange bias, i.e., the exchange coupling at the AF/F interface may depend sensitively on exactly its cooling history. Reproducible exchange coupling is required for practical devices, and should be further investigated in our LSMO heterostructures.

The main panel in Fig. 2 shows the temperature dependence of $TMR_{\text{max}}$, which decays quickly with increasing $T$ and vanishes at $\sim280$ K, which is also near the temperature at which exchange bias effects disappear. The strong decay of $TMR_{\text{max}}(T)$, could in principle be explained by different $T$-dependent mechanisms, for example, intrinsic to the LSMO/STO interfaces (such as intrinsic loss of spin polarization) or extrinsic (such as weakening of the exchange bias pinning of the bottom electrode). Any study of the temperature dependence of the domain structure and thus relative magnetization orientations of the electrodes would require a microscopic technique such as demonstrated in Ref. 23. Evidence for weakened exchange bias pinning at higher $T$ is seen in the decreasing asymmetry of $\Delta H$ and $R_{\text{max}}$ as temperature is increased; the asymmetry in $R(H)$ between positive and negative $H$ disappears around $\sim100$ K. Further characterization is necessary to distinguish between these competing mechanisms, and will be crucial to understand any limits to the potential high-temperature TMR.

Figure 3 shows a polar plot of $TMR_{\text{max}}(\alpha)$ at $T=30$ K, after field-cooling along the $a$-axis of LSMO, which still shows a slight asymmetry for opposite field directions. The fourfold symmetry of $TMR_{\text{max}}(\alpha)$ indicates a biaxial anisotropy with easy axis along the $a$- and $b$-directions of our F-LSMO layers. The slight difference in $TMR_{\text{max}}$ values for orientations close to the $a$- and $b$-axes could be due to, e.g., a shape effect correlated with the junction long axis, or a small anisotropy in the exchange bias; fourfold symmetry in the switching fields has been previously reported. 24 We note that cooling the device with the field oriented in different directions does not change the fourfold symmetry seen in the polar plot, although it can have a sizable impact on $TMR_{\text{max}}$.

Additional study of the interplay between junction geometry and magnetocrystalline anisotropy will be necessary to further optimize these MTJs.
In summary, we have shown $R(H)$ at 4 K for a manganite MTJ with a useful $TMR_{\text{max}}$ ratio of 1900%, the largest value for any MTJ reported so far in the literature at low dc bias. The strength of pinning of one electrode magnetization via exchange bias has a noticeable influence on the TMR but is also very sensitive to the cooling history. The polar plot of $TMR_{\text{max}}(\alpha)$ demonstrates that uniaxial anisotropy in the F layers is not necessary for high TMR. It is reasonable that interface roughness, oxygen vacancies and the interface growth play a crucial role in the exchange bias mechanism in these manganite interfaces as has been demonstrated in more conventional exchange bias systems, and merits further study.

R.W. gratefully acknowledges support by the Cusanuswerk, Bischöfliche Studienförderung. B.A.D. and A.Y.P. acknowledge support by the FVG Regional project SPLITNOX funded by Legge Regionale Grant No. 26/2005 and Decreto Grant No. 2007/LAVFOR/1461. L.A.-M. thanks the Abdus Salam ICTP (Trieste) for financial support through a STEP fellowship. The TASC Technical group is acknowledged for contributions to the design and construction of the MBE system. This work was funded in part by the Deutsche Forschungsgemeinschaft (Project No. KO 1303/8-1).

1S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science 294, 1488 (2001).
2M. Julliere, Phys. Lett. A 54, 225 (1975).
3J. M. De Teresa, A. Barthélémy, A. Fert, J. P. Contour, F. Montaigne, and P. Seneor, Science 286, 507 (1999).
4M. Bowen, J.-L. Maurice, A. Barthélémy, M. Bibes, D. Imhoff, V. Bellini, R. Bertacco, D. Wortmann, P. Seneor, E. Jacquet, A. Vaures, J. Humbert, J.-P. Contour, C. Colliex, S. Blugel, and P. H. Dederichs, J. Phys.: Condens. Matter 19, 315208 (2007).
5J. Z. Sun, D. W. Abraham, R. A. Rao, and C. B. Eom, Appl. Phys. Lett. 74, 3017 (1999).
6M. Bibes, L. Balcells, S. Valencia, J. Fontcuberta, M. Wojcik, E. Jedryka, and S. Nadolski, Phys. Rev. Lett. 87, 067210 (2001).
7A. Tebano, C. Aruta, S. Sanna, P. G. Medaglia, G. Balestrino, A. A. Sidorenko, R. D. Renzi, G. Ghiringhelli, L. Braicovich, V. Bisogni, and N. B. Brookes, Phys. Rev. Lett. 100, 137401 (2008).
8A. Verna, B. A. Davidson, Y. Szeto, A. Y. Petrov, A. Mirone, A. Giglia, N. Mahne, and S. Nannarone, J. Magn. Magn. Mater. 322, 1212 (2010).
9M. Viret, M. Drouet, J. Nassar, J. P. Contour, C. Fermon, and A. Fert, Europhys. Lett. 39, 545 (1997).
10M.-H. Jo, N. D. Mathur, N. K. Todd, and M. G. Blamire, Phys. Rev. B 61, R14905 (2000).
11M.-H. Jo, N. D. Mathur, N. K. Todd, and M. G. Blamire, Phys. Rev. B 61, R14905 (2000).
12J. O’Donnell, A. E. Andrus, S. Oh, E. V. Colla, and J. N. Eckstein, Appl. Phys. Lett. 76, 1914 (2000).
13H. Yamada, Y. Ogawa, Y. Ishii, H. Sato, M. Kawasaki, H. Akoh, and Y. Tokura, Science 305, 646 (2004).
14J. J. Kavich, M. P. Warusawithana, J. W. Freeland, P. Ryan, X. Zhai, R. H. Kodama, and J. N. Eckstein, Phys. Rev. B 76, 014410 (2007).
15M. Bowen, M. Bibes, A. Barthélémy, J.-P. Contour, A. Anane, Y. Lemaitre, and A. Fert, Appl. Phys. Lett. 82, 233 (2003).
16I. K. Schuller, Mater. Res. Bull. 29, 642 (2004).
17J. N. Eckstein and I. Bozovic, Annu. Rev. Mater. Sci. 25, 679 (1995).
18J. M. Haeni, C. D. Theis, and D. G. Schlom, J. Electroceram. 8, 385 (2000).
19B. A. Davidson, A. Y. Petrov, A. Verna, X. Torrelles, M. Pedio, A. Cosaro, and S. Nannarone (unpublished).
20See supplementary material at http://dx.doi.org/10.1063/1.3581885 for details on MTJ patterning.
21R. J. M. van de Veerdonk, J. Nowak, R. Meservey, J. S. Moodera, and W. Kodama, J. Appl. Phys. Lett. 71, 2839 (1997).
22M. Wagenknecht, H. Eitel, T. Nachtrag, J. B. Philipp, R. Gross, R. Kleiner, and D. Koelle, Phys. Rev. Lett. 96, 047203 (2006).
23M.-H. Jo, N. D. Mathur, and M. G. Blamire, Appl. Phys. Lett. 80, 2722 (2002).
24W. Kuch, L. I. Chelaru, F. Offi, J. Wang, M. Kotsugi, and J. Kirschner, Nature Mater. 5, 128 (2006).