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


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We report resistance versus magnetic field measurements for a La$_{0.65}$Sr$_{0.35}$MnO$_3$/SrTiO$_3$/La$_{0.65}$Sr$_{0.35}$MnO$_3$ 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 manganite 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. An MTJ consists of two ferromagnetic electrodes separated by a thin insulating tunneling barrier. According to the Julliere model, the TMR ratio is defined as $\text{TMR} = (R_{\text{AP}} - R_{\text{P}}) / R_{\text{P}} = 2P_1P_2 / (1 - P_1P_2)$. Here $P_1$ and $P_2$ are the spin polarizations of the two electrodes and $R_{\text{AP}}$ and $R_{\text{P}}$ are the junction resistances with antiparallel and parallel orientation of magnetization $M$ of the two electrodes, respectively. Accordingly, an MTJ made from half-metallic electrode materials, such as doped manganites, 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, this concept has been extended to also describe interfaces as half-metallic, 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 manganite surfaces and interfaces are known to be weaker than in bulk, causing a “dead layer.” For example, at the vacuum/La$_{1-x}$Sr$_x$MnO$_3$ (LSMO) interface the nonferromagnetic layer is about three unit cells (uc) thick at $T=200$ K, 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 manganite MTJs. 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, spectroscopic characterization suggested this approach could improve the low-temperature TMR.

To date, TMR at small dc voltage bias of MTJs based on nonoxide electrodes reached $\approx 1150\%$ at $T=5$ K (Ref. 15) while the highest ratio was reported for manganite/titanate interfaces with a maximum value of about 1800% at 4 K in a very small window of applied in-plane magnetic field $H$. Here, an antiferromagnetic CoO layer was used to pin the upper electrode via exchange bias 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.

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 SrTiO$_3$ (STO) barrier, grown by molecular beam epitaxy (MBE). We find a TMR ratio up to $\approx 1900\%$ at $T=4$ K, which decreases rapidly with increasing $T$, disappearing at $\approx 280$ 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. These RHEED techniques permit us to adjust the surface termination at any point during deposition, including during interface growth. 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 stoichiomic 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 $\approx 250$ K, in agreement with exchange bias effects seen in the hysteresis loops of an AF-LSMO (100 uc)/F-LSMO (50 uc) junction.
Few nanoampere. Full characterization of the junction resistance was measured independently. MTJs with a 5 μm² mesa with vertical current injection allows full rotation of the crystallographic lattice. The main panel in Fig. 2 shows the temperature dependence of TMRmax, which decays quickly with increasing T and vanishes at ~280 K, which is also near the temperature at which exchange bias effects disappear. The strong decay of TMRmax(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 ΔH and Rmax as temperature is increased; the asymmetry in R(H) between positive and negative H disappears around ~100 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 TMRmax(α) 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 TMRmax(α) indicates a biaxial anisotropy with easy axis along the a- and b-directions of our F-LSMO layers. The slight difference in TMRmax 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. 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 TMRmax. 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 manganese 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 both by enlarging the magnetic field window of antiparallel alignments and inducing an asymmetry in the $R(H)$ curves 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 manganese interfaces as has been demonstrated in more conventional exchange bias systems, and merits further study.

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FIG. 3. (Color online) Polar plot of the maximum TMR ratio (at $T=30$ K) vs field direction $\alpha$ for field-cooling at $\alpha_{cc}=70^\circ$ (dashed arrow). Crystalline axis are indicated by solid arrows; junction orientation is indicated by rectangle.