

Resistive Switching in All-Oxide Ferroelectric Tunnel Junctions with Ionic Interfaces

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Ferroelectric tunnel junctions have opened up promising routes towards energy-efficient data storage applications and memristive devices [1]. Polarization reversal in a ferroelectric tunnel barrier can change the electrical resistance of a junction, a phenomenon known as tunneling electroresistance (TER). On the other hand, redox-based effects such as the migration of oxygen vacancies can cause large resistive switching in transition metal oxides [2]. Here, we demonstrate nearly identical switching behavior in nominally symmetric tunnel junctions that are comprised of two La_{2/3}Sr_{1/3}MnO₃ (LSMO) electrodes separated by a ferroelectric PbZr_{0.2}Ti_{0.8}O₃ (PZT) or BaTiO₃ (BTO) tunnel barrier, or a paraelectric SrTiO₃ (STO) tunnel barrier [3]. Giant changes in electrical resistance with RH/RL ratios of up to 10⁶ are measured on 20 nm LSMO/2.5 nm barrier/20 nm LSMO junctions, whereas the effect is substantially reduced for thicker barriers. The invariance of resistive switching with barrier material and its anomalous dependence on tunnel barrier thickness strongly point towards a redox-based effect that is not noticeably influenced by ferroelectric polarization in the barrier. From transmission electron microscopy measurements, it can be found that the bottom LSMO/barrier interface is atomically sharp, but that the top barrier/LSMO interface extends over three unit cells. Current-voltage characteristic and data fits based on models for direct tunneling and Fowler-Nordheim tunneling indicate that the redox-based mechanism leads to the formation of an insulating layer in the LSMO bottom electrode when a positive bias voltage is applied to the top electrode. This conclusion is independently verified by in-plane electrical transport measurements on Hall bar structures. The following physical picture emerges from the experimental data: At positive bias voltage, oxygen vacancies migrate into the bottom LSMO electrode. This changes the valency of the Mn ions and enhances the electrical resistance. If enough oxygen vacancies migrate into the bottom electrode, a thin layer of LSMO next to the barrier becomes insulating, which leads to a giant increase of the junction resistance. Under negative bias voltage, the oxygen vacancies migrate back and the low resistance state is re-established. A similar sequence of events does not take place at the top LSMO/barrier interface because oxygen vacancy migration is hampered by structural roughness and

atomic mixing. Electric-field induced oxygen migration is reduced for thicker tunnel barriers and this limits the resistive switching effect.