Space-charge trap mediated conductance blockade in tunnel junctions with half-metallic electrodes

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A conductance blockade effect has been observed in the magnetic tunnel junction consisting of La₀.₆₇Sr₀.₃₃MnO₃ electrodes and a SrTiO₃ barrier. The blockade effect is correlated with the space-charge trap states in the barrier. The blockade threshold eVₜ₁=128 meV is significantly greater than Coulomb charging energy Eₑᵣ=11 meV. The blockade can be lifted with a magnetic field, accompanied by a very large magnetoresistance up to 10 000%. The intriguing blockade behavior is distinctly different from the conventional Coulomb blockade effect, showing a unique spin-dependent tunneling process mediated by the localized charge trap states. © 2008 American Institute of Physics. [DOI: 10.1063/1.3025851]

One of the main characteristics of few-electron systems with reduced dimensions is the Coulomb blockade effect, the suppression of current flow when the energy cost of introducing an extra electron in the system is large due to the Coulomb repulsion. If the spin of the electron comes into play, new phenomena emerge resulting from the interplay between spin-dependent transport and single-electron charging effects.¹ A striking example is the oscillations of tunneling magnetoresistance (TMR) as a function of bias voltage in Al/Al–O/Co–Al/Co granular films/Co/Pt multilayers that are associated with the discrete energy levels and large charging energies in Co nanoparticles.² The potential of manipulating spins one by one would open a promising path to quantum computing. Therefore, there is a growing interest to couple ferromagnetic electrodes to small objects in order to study spintronics in reduced dimensions. Some early attempts have already been encouraging, e.g., a TMR enhancement by 40% was obtained in a series of Ni/NiO/Co small magnetic tunneling junctions (MJTs) in the Coulomb blockade regime.³ Even higher TMR enhancement was predicted at very low temperature in the strong tunneling regime by taking into account higher order tunneling processes.⁴ A TMR of more than 1800% was reported in the La₀.₆₇Sr₀.₃₃MnO₃/SrTiO₃/La₀.₆₇Sr₀.₃₃MnO₃ MTJ.⁵ This is because the LaₓSrₓMnO₃ (LSMO) electrode with x=0.3 has nearly total spin polarization, namely, nearly half-metallic.⁶,⁷ In this letter, we report a conductance blockade in the LSMO/STO/LSMO MTJ, in which the tunneling process is dictated by localized charge trap states in the STO barrier layer. A very large TMR has been observed in association with a unique interplay of half-metallicity of electrode and the single-electron tunneling process at the charge trap states.

LSMO (100 nm)/STO (3 nm)/LSMO (100 nm) epitaxial trilayer structures were grown onto (001)-oriented SrTiO₃ substrates using rf magnetron sputtering. Both LSMO and STO layers were deposited at 700 °C in a mixture of Ar and O₂ (4:1) under a pressure of 1.0 Pa. The trilayer thin films were then annealed in situ at the growth temperature for 30 min in an O₃ atmosphere. This growth and annealing process has been shown to reduce the oxygen vacancy density. A platinum layer (20 nm thick) was then deposited ex situ by dc magnetron sputtering on top of the trilayer to protect the top LSMO surface from possible damage. Ellipscus-pillar-like microscale junctions (3×6 μm²) were fabricated using a conventional microfabrication technique.⁸ The transport behaviors were measured with a four-probe method using a Keithley 6430 subfemtoampere electrometer in a cryogenic environment.

The heterojunction quality was examined using cross-sectional high-resolution transmission electron microscopy (HRTEM) and the energy dispersive spectroscopy (EDS). Figure 1 shows the HRTEM image with atomically sharp interfaces and the energy dispersive spectroscopy (EDS) profiles across the heterojunction. The STO layer is well defined with the coincident Sr peak and the Mn minimum. The Ti line is overwhelmed by background noise and no peak is visible.

The transport properties of MTJs show a sample-to-sample variation. Two junctions presented here are, junction 1 that displays a conductance blockade and junction 3 that does not. The differential conductance dI/dV of a MTJ (junction 1) is shown in Fig. 2. At temperatures higher than 100 K, the conductance remains finite at all bias voltages [Fig. 2(a)], namely, there is no blockade. However, at lower temperatures, a strong suppression of conductance is seen at low bias range [Fig. 2(b)], showing a clear conductance blockade. The conductance blockade threshold is strongly temperature dependent, and a magnetic field can lift the blockade, as shown in Fig. 2(b) at T=10 K.

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The conductance blockade effect is shown to correlate with the charge trap states in STO barrier layer. Figure 4 shows the temperature dependence of the zero bias conductance $G_0 = dI/dV(V=0)$ for junctions 1 and 3. The conductance of junction 1 follows the Arrhenius form $G_0(T) = A \exp(-E_a/kT)$. The fitting parameter $E_a=67$ meV is interpreted as the ionization energy of the dominant space-charge trap level. The charge trap states can be attributed to the existence of the oxygen vacancy, a fundamental and intrinsic defect in perovskite oxide barrier layer. The effective positive potential of the oxygen vacancy alters the electronic properties of STO, leading to the formation of localized electron trapping states within the energy gap. The conductance of junction 3, however, is not thermally activated and no blockade has been observed.

In the presence of space charges, the conductance becomes space-charge limited when the electronic screening effect in the barrier layer reaches to the point to block further current flow at steady state. Actually, a space-charge limited tunneling was observed in LSMO/STO/LSMO MTJs. In the space-charge limited conductance regime, the Coulomb charging effect will appear when the total space charge is on the order of one electron. In Arrhenius plot of Fig. 4(a), the fitting parameter $A=2.2 \times 10^{-4} \Omega^{-1}$ and thus $A/(2e^2/h)=3$, corresponding to the extracted number of effective conduction channels in the junction; indeed, this is a few-electron tunneling system. Note, the number of actual defect in the junction should be much higher than that of effective conduction channels as many defect states may not be active in
transport at this temperature. Thus, conductance oscillations appear in Figs. 2(b) and 3 can be attributed to the Coulomb charging effect. The oscillation period is approximately 22 mV, corresponding to charging energy $E_C = 11$ meV.

The conductance blockade effect can be lifted by the magnetic field. Figure 4(b) shows the tunneling current $I$ at three different voltages, 50, 100, and 150 mV, as a function of the magnetic field $H$ at $T=10$ K. 150 mV is above the blockade voltage while the other two values are below. For 50 and 100 mV, the conductance remains blocked when $\mu_0H<5$ T. The current then increases linearly with the field when $\mu_0H>5$ T. For 150 mV, where there is no longer a blockade effect, the current grows exponentially with the field. This leads to a very large TMR at high fields without any indication of saturation. The TMR are shown in the inset of Fig. 4(b) for junction 1. Here the TMR is defined as $[R(0)-R(H)]/R(0)$ where $R(0)$ is the zero field resistance and $R(H)$ is the resistance under magnetic field $H$. The highest TMR is close to 10 000% at $\mu_0H=14$ T. For junction 3 that does not display a blockade, a lower TMR of 1920% has been obtained. This TMR value confirms the nearly total spin polarization of the electrodes. Thus, the unusually high TMR is a consequence of the blockade effect. It is worth noting that the activation energy of dominant trap states and the density of the traps depend critically on the material growth condition and device fabrication process. Therefore, a big sample-to-sample conductance variation appears as displayed by junctions 1 and 3. Similar variations have also been observed in defect state mediated resonant tunneling processes such as cotunneling can lead to an enhanced TMR compared to that in the sequential regime. However, this enhancement is expected to disappear when sequential tunneling is restored at $V_{dc} > V_B$. A huge TMR exists even beyond the blockade voltage cannot be explained with a cotunneling mechanism. Simple spin-dependent scattering due to spin disorder at LSMO/STO interfaces cannot produce TMR of this magnitude either, and would also saturate at much lower magnetic fields. The unusual blockade effect and the accompanying high TMR are a unique consequence of interplay of half-metallic nature of LSMO electrode and the single-electron tunneling process at the charge trap states in STO. The underlying mechanism is to be explored.

In summary, a space-charge trap induced conductance blockade effect is observed in LSMO/STO/LSMO tunnel junctions. A large blockade threshold has been observed that is significantly higher than Coulomb charging energy. The blockade effect can be lifted by a magnetic field, accompanied by a very large TMR at a voltage beyond blockade threshold. The unusual blockade effect and very large TMR value cannot be reconciled with the concept of conventional Coulomb blockade, indicating a unique consequence of spin-dependent transport in single-electron tunneling regime.

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