Inversion of Magnetoresistance in Magnetic Tunnel Junctions: Effect of Pinhole Nanocontacts

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Inverse magnetoresistance has been observed in magnetic tunnel junctions with pinhole nanocontacts over a broad temperature range. The tunnel magnetoresistance undergoes a change of sign at higher bias and temperature. This phenomenon is attributed to the competition between the spin conserved ballistic transport through the pinhole contact where the transmission probability is close to unity and spin polarized tunneling across the insulating spacer with weak transmittivity.

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During the last decade, the study of spin polarized tunneling in magnetic tunnel junctions (MTJs) [1] has experienced an exponential growth. Recently, the theoretical prediction and subsequent observation of large room temperature magnetoresistance in epitaxial Fe/MgO/Fe structure [2] have generated tremendous interest among physicists from both fundamental and technological points of view. However, the influence of ballistic spin dependent transport (due to the presence of pinhole nanocontacts which connect the two ferromagnetic electrodes) on the magnetoresistive properties of MTJs has not been explored substantially. Recent simulations have shown that as much as 88% of the current can flow through the pinholes [3] in MTJs even though the bias dependence of differential conductance has a positive curvature. There are reports of large ballistic magnetoresistance (BMR) at room temperature [4] in ferromagnetic nanocontacts. The essential ingredient for BMR is the condition of nonadiabaticity in ballistic transport across the nanocontact. If the domain wall width at the nanocontact is sufficiently thin so that the spin does not have time to flip then the situation becomes analogous to the spin conserved tunneling in MTJs [5]. In that case the BMR is related to the spin polarization of the electrodes in the same way as the tunneling magnetoresistance (TMR) in Julliere [6] or Slonczewski's model [7] which predicts positive TMR for symmetric electrode MTJ. However, it is claimed [8] that ballistic channels in MTJs are not magnetoresistive and the opening up of a spin-independent conduction channel can only reduce the TMR. We will show that the ballistic channel in MTJs are not only magnetoresistive; it, in fact, can cause inverse tunneling magnetoresistance. The relative contributions from the two conduction channels—elastic tunneling through the insulating spacer and ballistic spin polarized transport through the narrow pinhole shorts—can change as the temperature and applied bias are varied and magnetoresistive response can change accordingly.

Observation of inverse TMR where the conductance in the antiparallel magnetic configuration is higher than that in the parallel configuration, has been instrumental in understanding some of the important aspects of spin polarized transport in MTJs. For example, the inverse TMR observed in experiments by De Teresa *et al.* [9] have proved that the transport properties of MTJ depend not only on the ferromagnetic metal electrodes but also on the insulator. Generally, inverse TMR can occur if the sign of spin polarization of the two electrodes is opposite in the relevant bias range.

An interesting inversion of TMR has been observed by us in a broad temperature range. The observed inverse TMR ($\mathrm{TMR} = \delta R/R = (R_{\mathrm{AP}} - R_{\mathrm{P}})/R_{\mathrm{P}}$ where R_{AP}, R_{P} are the junction resistances in antiparallel and parallel magnetic configuration of the MTJs, respectively) changes sign as the bias voltage and temperature is increased. The inverse TMR is attributed to the spin conserved transport through nanoscale metallic channel where the transmission probability is close to unity. Spin polarized tunneling with weak transmission probability dominates at higher temperature and bias leading to normal positive TMR.

The trilayer $\mathrm{La_{0.67}Sr_{0.33}MnO_3}$(LSMO)/$\mathrm{Ba_2LaNbO_6}$(BLNO)/LSMO was deposited on single crystalline $\mathrm{SrTiO_3}$ (100) substrate held at a temperature 800 °C and oxygen pressure 400 mTorr, using pulsed laser deposition. Thickness of the bottom LSMO layer is 1000 Å and that of the top layer 500 Å while the estimated thickness of the insulating spacer from the deposition rate calibration of BLNO is 50 Å. The microfabrication was done using photolithography and ion-beam milling. For details see Ref. [10].

There is a set of criteria, known as Rowell's criteria [11], for determining the quality of the tunnel junction. However, for magnetic tunnel junctions, only three of these criteria are applicable: (1) Exponential thickness dependence of junction resistance. (2) Parabolic differential conductance curves that should be well fitted by rectangular barrier Simmons [12] model or trapezoidal barrier Brinkman model [13]. (3) Insulating like temperature dependence of junction resistance. It has been observed that MTJs with pinhole shorts can reproduce the first two criteria [14]. Therefore the third criteria stands out as the reliable proof of the quality of the MTJ. Although the junctions show nonohmic voltage dependence, the temperature dependence of junction resistance is metal-like [Fig. 1(a)]. In this Letter, we will show that two MTJs with pinhole shorts exhibit almost identical magnetoresistive properties although the voltage dependence of differential conductance curves have opposite curvatures. While the sample denoted MTJ1 shows positive curvature in the conductance curve, the conductance of MTJ2 has negative curvature [Fig. 1(b)]. The MTJs contain metallic nanocontacts through which electrons travel ballistically at low temperature and bias. However, at higher bias, ''hot electron'' transport through the pinholes results in heat dissipation within the nanocontact region just outside the ballistic channel [15] and thus increases the resistance. At higher bias the backscattering into the narrow channel increases due to larger phonon density of states at the nanocontact, which reduces the transmittivity resulting in negative curvature in the voltage dependence of differential conductance. However, the conduction channel due to tunneling will become less resistive at higher bias since then the electrons will tunnel across the relatively thin trapezoidal part of the barrier. As a result, the pinhole short will produce negative curvature in the differential conductance curve while tunneling should cause positive curvature. Although transport in both the MTJs is dominated by conduction through pinhole shorts, which is evident in Fig. 1(a), the strong positive curvature in the voltage dependence of conductance due to tunneling can overcome the weak negative curvature due to transport through the pinholes, resulting in overall positive curvature as observed in MTJ1 [Fig. 1(b)]. Fitting the differential conductance curves with positive curvature for MTJ1 by the Brinkman model, the extracted barrier height turns out to be about 0.8–1 eV (much higher than the value 0.2–0.3 eV corresponding to MTJs without pinhole shorts) and the barrier width much smaller (15–20 Å) compared to that of ~40 Å for good MTJs. The extracted value for barrier height increases while the barrier width decreases as the temperature is increased. Although the value of the barrier parameters, in the present case, carry no physical significance, temperature dependence of the barrier parameters is a reconfirmation of the MTJ having pinhole shorts [16].

Inverse TMR is observed for both MTJs over a broad temperature range 10–150 K (Fig. 2). The value of inverse TMR decreases with increasing temperature. For MTJ1 the value of inverse TMR is 4.6% at 10 K which reduces to about 1.8% at 150 K, while for MTJ2 it is about 6.5% at 10 K which almost vanishes at 150 K. Above 150 K, the situation is the opposite—ordinary positive TMR is observed. At 200 K, the positive TMR exhibited by MTJ1 is about 1% while that for MTJ2 is 0.06% [Figs. 2(c) and 2(f)]. The bias dependence of TMR for MTJ1 has some interesting features. At 150 K, it is observed that above ±225 mV, the TMR changes sign [Fig. 3(a)]. A clear evidence of such inversion is highlighted in Figs. 3(c) and 3(d) where MTJ1 shows inverse TMR at bias current $I = 200\ \mu\mathrm{A}$, while at $I = 1$ mA it exhibits positive TMR. However, at a lower temperature 100 K, there is no evidence of such inversion with increasing bias [Fig. 3(b)].

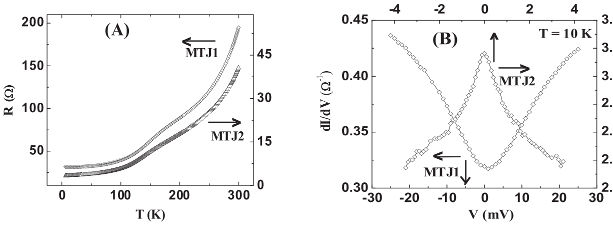

FIG. 1. (a) Junction resistance vs temperature curves for pinhole short MTJ1 and MTJ2 showing metal-like temperature dependence of resistance. (b) Nonohmic voltage dependence of differential conductance showing opposite curvatures for MTJ1 and MTJ2.

The observed phenomenon can be explained as follows. The present system can be considered as being equivalent to two ferromagnetic metal electrodes connected by ballistic nanoscale metallic channels along with a conduction channel connected in parallel which describes tunneling across the insulating spacer. For the case of two identical ferromagnets connected by a nanocontact, the BMR [5] is given by

$$\Delta R/R_{\mathrm{P}} = \frac{2P^2}{1 - P^2} f(k_F \lambda)$$

where P is the spin polarization, λ is the domain wall width, and k_F is the Fermi wave vector, f being the measure of the spin nonconservation in the current through

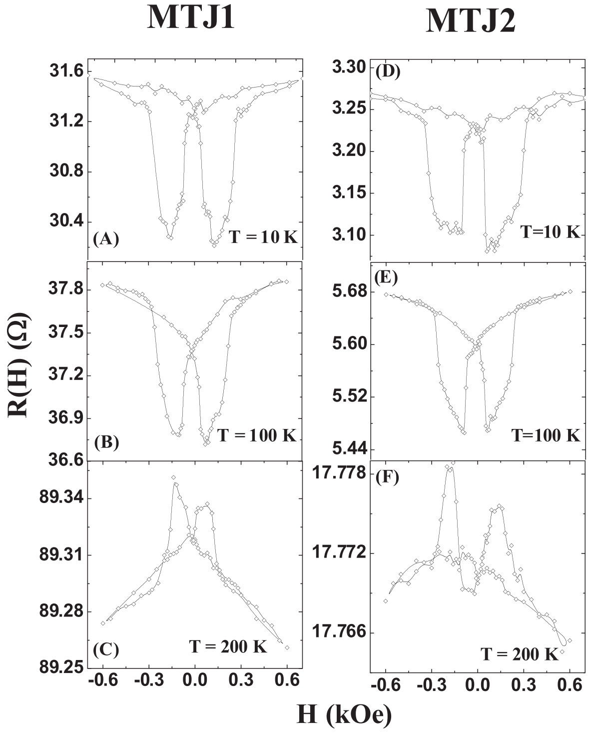

FIG. 2. Junction resistance vs magnetic field curves for pinhole short MTJ1 (a), (b), (c) and MTJ2 (d), (e), (f) at different temperatures showing the inverse TMR at low temperature which undergoes a change of sign as temperature is increased to 200 K.

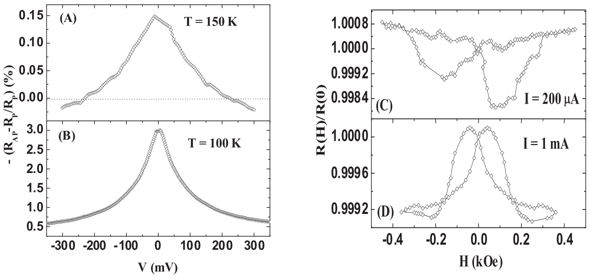

FIG. 3. (a) Bias dependence of TMR for MTJ1 at 150 K showing bias induced inversion of magnetoresistance above ±225 mV. (b) Bias dependence of TMR for MTJ1 at 100 K showing no evidence of sign change of TMR. The reduced junction resistance vs magnetic field curves for MTJ1 at 150 K at bias currents $I = 200\ \mu$A (c) and $I = 1$ mA (d). At low bias, inverse TMR is observed while at high bias the sign of TMR reverses resulting in positive TMR.

the nanocontact. In the limit of vanishing domain wall width λ, spin flipping by domain wall scattering is absent. Then f is unity and the electron spin is conserved during transmission (the factor f decreases with the increase of the product $k_F\lambda$). Hence we arrive at the well-known Julliere formula for tunneling magnetoresistance. Thus there seems to be no difference in the spin conserved ballistic transport through nanosized pinholes or elastic spin polarized tunneling. However, there is a stark contrast in the transmittivities for the two conduction channels. In case of normal elastic tunneling through the insulating barrier the tunneling probability is finite but small and decays exponentially with increasing barrier width. On the other hand, the transmittivity through the metallic pinhole nanocontact is close to unity.

At low temperature, the electron transfer from one ferromagnetic lead to another occurs dominantly through the metallic pinhole shorts. Hence, according to Ref. [5], the ballistic magnetoresistance should follow the Julliere or Slonczewski's model for spin polarized tunneling and should give positive TMR for MTJs with identical electrodes. However, in our case, inverse TMR is observed. The reason probably lies in the fact that the model does not take into account the effect of high transmittivity and the possibility of different transmission coefficients of the electrons in the majority and minority spin bands. The model reduces to the Julliere model in the nonadiabatic limit. However, there is a general agreement that the generalized Julliere model is valid in the limit of very weak transmission probability [17]. Kim [18] has very recently put forward a theoretical model for spin polarized transport through a narrow channel. In this treatment, when the spin is conserved in transport through a nanoscale channel and the transmittivity is close to unity, there is a possibility of inverse TMR. According to Kim's model, transmission probabilities in the parallel and antiparallel magnetic configuration of the two electrodes (assuming that the spin polarizations of the two electrodes are the same) are given as

$$T_{\mathrm{P}} = \frac{2\gamma_+}{(1+\gamma_+)^2} + \frac{2\gamma_-}{(1+\gamma_-)^2}, \qquad T_{\mathrm{AP}} = \frac{4\sqrt{\gamma_+\gamma_-}}{(1+\sqrt{\gamma_+\gamma_-})^2}, \tag{1}$$

where γ_+ and γ_- are the transfer rates for majority and minority spins, respectively. When the transmission probability is small, i.e., $\gamma_\pm \ll 1$, $T_{\mathrm{P}} = 2(\gamma_+ + \gamma_-)$, $T_{\mathrm{AP}} = 4\sqrt{\gamma_+\gamma_-}$, which means that the transmission probability in the parallel configuration is greater than that in the antiparallel configuration; i.e., the TMR is positive. The conditions for zero TMR or $T_{\mathrm{P}} = T_{\mathrm{AP}}$ are given as $\gamma_+ - \gamma_- = 0$, which is a trivial solution and implies that spin polarizations at the Fermi level for both the electrodes is zero and is applicable for nonmagnetic tunnel junctions. The nontrivial solution for zero TMR with spin polarization $P \neq 0$, resides at the boundary between two regions corresponding to $T_{\mathrm{P}} > T_{\mathrm{AP}}$ and $T_{\mathrm{P}} < T_{\mathrm{AP}}$ and is given by

$$(\gamma_+\gamma_- - 1)^2 - 2\sqrt{\gamma_+\gamma_-}(1+\gamma_+)(1+\gamma_-) = 0.$$

To be more precise, the combinations (γ_+, γ_-) satisfying the above equation constitutes a curve in (γ_+, γ_-) space enclosing the region where normal positive TMR occurs. The region outside the curve contains high values for γ_+ and γ_- which corresponds to inverse TMR. The transmission probabilities for the majority and minority spin band are related to $\gamma_\pm$ as follows:

$$T_\pm = \frac{4\gamma_\pm}{(1+\gamma_\pm)^2}.$$

Thus, when transmission probability is closer to unity, i.e., $T_\pm \simeq 1$, and there is an imbalance in the transmission probabilities for the majority spin and the minority spin, inverse TMR occurs.

Replacing $\gamma_\pm$ by $T_\pm$ in the expression for T_{P} and T_{AP}, the TMR values ($\Delta R/R_{\mathrm{P}} = \{T_{\mathrm{P}} - T_{\mathrm{AP}}\}/T_{\mathrm{AP}}$) can be calculated for all possible values of $T_\pm$. The theoretically allowed values of (T_+, T_-) for inverse TMR and how the allowed values of (T_+, T_-) evolve with the change in temperature for MTJ1, within the bias range ±220 mV, are shown in Fig. 4 along with the corresponding TMR values. The contribution due to the parallel tunneling conduction channel has been neglected for simplicity of calculation. This will, of course, lead to underestimation of the allowed values of $T_\pm$, particularly in the high temperature region where the relative contribution of the tunneling conduction channel will be substantial. The calculation suggests that, the larger the imbalance between T_+ and T_-, the greater is the value of inverse TMR as can be seen from Fig. 4(a). Up to 100 K, the allowed values of $T_\pm$ stay away from the $T_+ = T_-$ line [Figs. 4(b) and 4(c)]. However, as the temperature is increased further, the imbalance in the transfer rates of majority and minority spins diminishes drastically and the allowed values congregate near $T_+ = T_-$ [Fig. 4(d)]. The increase in bias also reduces the imbalance between the transmittivities in the two bands

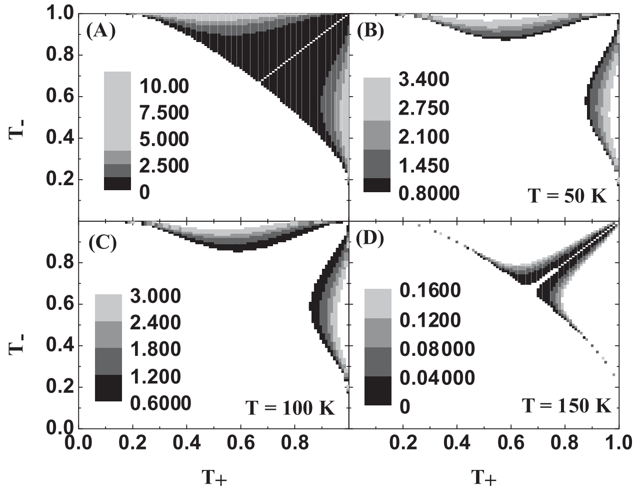

FIG. 4. (a) The theoretically allowed values of $T_{\pm}$ for inverse TMR in the $\{T_+, T_-\}$ plane and the corresponding values of inverse TMR is shown by a shaded map. (b), (c), (d) The allowed values of $T_{\pm}$ which causes inverse TMR for MTJ1 at 50, 100, and 150 K, respectively, within the bias range ±220 mV. The corresponding values of inverse TMR in each case is shown by the shaded map.

as can be seen from the color map for each temperature. If the values of T_+ and T_- are interchanged the TMR remains the same. However, the physically acceptable situation is where T_+ is greater than T_-, since the minority spin states are generally regarded as being more localized compared to the majority spin states.

Although $La_{0.67}Sr_{0.33}MnO_3$ is generally considered to be having almost full spin polarization, Andreev reflection experiments have confirmed the existence of minority spin states which will be particularly influential in the ballistic limit of transport [19]. The change from inverse TMR to a positive one at higher bias at 150 K can be attributed to the fact that at higher bias electrons tunnel through relatively thin trapezoidal part of the barrier such that the contribution due to elastic tunneling increases which gives rise to positive TMR. On the other hand, there are several reasons for the decrease of inverse TMR at higher bias due to transport through the narrow channel. (1) Local generation of heat within the nanocontact region at higher bias leads to increased thermal spin fluctuation and resistance at the nanocontact which reduces the inverse TMR. (2) The backscattering into the narrow channel increases as a result of larger phonon density of states at the nanocontact, reducing the transmittivity and hence the inverse TMR. (3) Lastly, the product $k_F\lambda$ being larger at higher bias may cause deviation from the nonadiabatic limit. This results in increased spin flip scattering thus reducing the magnitude of inverse TMR. In our case, the normal positive TMR is observed at 200 K, where elastic tunneling across the insulating spacer with weak tunneling probability is dominant and the electron-phonon interaction is large enough to push the transport through the pinhole into diffusive regime. Manganese oxide tunnel junctions with pinhole shorts are better suited to exhibiting inverse TMR than MTJs with transition metal electrodes since in that case there is a high probability of the pinhole shorts getting oxidized, which would lead to weak transmittivity through the narrow channel.

To summarize, we have presented direct experimental evidence that pinhole shorts through the insulating spacer in a magnetic tunnel junction can cause inverse tunneling magnetoresistance when the transmission probability is close to unity, which is an indicator that the Julliere and Slonczewski models are no longer valid in this regime. The relative contributions from the conduction channels due to elastic tunneling and ballistic spin conserved transport through the pinholes can be changed by proper adjustment of the bias and temperature, which can even result in the change of sign of the tunneling magnetoresistance.

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