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Voltage-induced switching with magnetoresistance signature in magnetic nano-filaments

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Abstract

Large hysteretic resistance changes are reported on sub-100 nm diameter metallic nanowires including thin dielectric junctions. Bi-stable 50% switching in a double junction geometry is modeled in terms of an occupation-driven metal–insulator transition in one of the two junctions, using the generalized Poisson expressions of Oka and Nagaosa (2005 \textit{Phys. Rev. Lett.} \textbf{95} 266403). It illustrates how a band bending scheme can be generalized for strongly correlated electron systems. The magnetic constituents of the nanowires provide a magnetoresistive signature of the two resistance states, confirming our model and enabling a four states device application.

((Some figures in this article are in colour only in the electronic version)
Figure 1. (a) Sketch of the double junction nanowire, involving successive growth of Ni (bottom electrode), NiO, (first junction), Co followed by Ni (island), NiO, (second junction), and Co (top electrode). Transmission electron micrograph (TEM) of the samples. (b) Ni–NiO–(CoNi), with a typical size of the CoNi island. (c) Ni–NiO–NiCo–NiO–Co nanowire with the central island showing very low contrast.

were used as templates. The metallic segments of nanowires filling the membrane were deposited in sequential order by replacing the bath containing Ni and Co ions, and applying an adequate deposition potential. Smooth and uniform NiO is obtained by anodization of the Ni top layers in 0.075 M Na3BO3 and 0.3 M H2B4O7. Characterization of the dielectric layer properties was made in situ by means of impedance spectroscopy. The estimated thickness is found to be about 1.5 nm [17]. Mott–Schottky analysis reveals the presence of p-type impurities with a concentration about \( N_d = 10^{25} \) m\(^{-3}\), somewhat lower than previously reported [18]. For TEM observation purposes, membranes filled with nanowires were dissolved in order to perform TEM analysis of the samples. The diameters of the nanowires were found to be in the range of 70 ± 20 nm, significantly larger than the nominal size of the membrane pores [19].

Nickel oxide is a Mott-type insulator that exhibits electroresistive switching properties [9, 11], often interpreted with a filamentary conduction model [20]. Our previous results on TMR single junctions in Ni/NiO/Co nanowires showed that filamentary conduction indeed occurred in those very small junctions [15]. Using tools developed for performing electrical connections to a single wire [21] we ensured that the investigated samples had sizes below 100 nm in diameter. Further reduction results in hot spots for the current passing through the NiO oxide. We previously observed that transport occurs within an area limited to a hot spot of a few nanometers size, for which single impurities were controlling the transport properties [14]. Extensive statistical studies indicated that impurity states in the barrier provide resonant tunneling paths, significantly affecting the TMR properties. The most significant outcome is the possibility to reverse the TMR sign [15].

No indications of resistance switching were found in more than 200 Ni/NiO/Co single junctions samples, possibly due to their limited voltage stability, ultimately resulting in irreversible breakdown under a voltage bias typically exceeding 100 mV. We extended our study to double insulating barrier heterostructures Ni–NiO–(CoNi)–NiO–Co, obtained by incorporating a CoNi island by electrodeposition and repeating the anodization process for NiO fabrication. This middle segment was made by consecutive Co and Ni deposition, where Co plating from non-aqueous solution was necessary to keep the integrity of the insulating barrier, and Ni coating was essential for repeating the anodization process. Systematic electron microscopy imaging (figure 1) provided calibration of the thickness of the deposit as a function of the electric charge passed between working and counter-electrode. The smallest Co–Ni thickness was 50 nm, and all the results presented here are for islands of sizes between 50 and 150 nm, which is the typical spread of the observed thickness under TEM observation. Samples made of longer islands showed results corresponding essentially to those observed for single junctions.

Hysteretic switching of the resistance value was observed at temperatures below 20 K on heterostructures with double NiO junctions (figure 2). The current value abruptly and irreversibly increases when the applied voltage reaches a threshold value of a few tens of mV, at the onset of the low resistance (LR) state. The opposite potential of similar magnitude switches the system back to the high resistance (HR) state. The change of resistance values spanned magnitudes between 2% and 100%, found on 26 samples (representing 20% success rate, also limited by breaking during cool-down or voltage sweeps). Nine samples showed reproducible behavior for more than ten voltage sweeps, without significant changes in the temperature range 1.5–20 K. The magnetoresistance properties of the two resistance states (LR, HR) can be remarkably different, and are reminiscent of those found on single junction devices. The example of
at 30 mV, of the two resistance states, with the color coding
switching. Bottom: magnetoresistance curves, under a DC bias held
barrier nanowire, measured at 1.5 K, showing reversible resistance
change. The change of sign between the two
signs as well as the applied magnetic field values triggering
to discard heating effects.

order of magnitude larger than those reported here, allowing us
thermometry measurements [24]. The deduced heating of the
induced magnetization reversal mechanism [22] is unlikely
switching between HR and LR states. A model of current-
figure 2 exhibits drastic differences in TMR properties when
switching between HR and LR states. A model of current-
induced magnetization reversal mechanism [22] is unlikely
to occur at the limited current densities we use, and cannot
explain the data, since the magnetic-field-induced change has a
sign and amplitude not corresponding to the voltage-induced
change. Joule heating triggering the resistance switching was
considered by several authors [4, 11, 23]. We took advantage
of the sporadic occurrence of telegraph noise in single junction
deVICES under significant potential bias [14] to perform local
thermometry measurements [24]. The deduced heating of the
junction did not exceed 10 K for current voltage products one
order of magnitude larger than those reported here, allowing us
to discard heating effects.

TMR properties of the HR and LR states differ in
signs as well as the applied magnetic field values triggering
resistance changes. The change of sign between the two
magnetoresistance curves of figure 2 can be interpreted in
terms of a formation/annihilation of a resonant state, providing
a new preferred path for the electron to flow. We propose that
one of the two junctions becomes conductive under an applied
bias by means of the filling-driven Mott–Hubbard metal
insulator (MI) transition model. This assumption explains the
abruptness of the transition and its hysteretic nature. While
stoichiometric nickel oxide with Ni^{2+} is an insulator, NiO,
with a large density of valence states can be brought very
close to the metal–insulator transition. The Ni electrochemical
anodization process is known to induce Ni^{2+}, Ni^{3+} and even
Ni^{4+} along with nickel vacancy states.

The occurrence of the MI transition in one of the barriers
can be explained semi-quantitatively by using a simple carrier
depletion/injection model. Figure 3 provides an intuitive
qualitative picture of the process. When putting the p-type
semiconductor NiO into equilibrium with the electrodes, a
hole-rich region (figure 3(b), left) appears at the positive
metal/electrode interface and a hole-deficient region near the
island promotes the insulating state. The applied voltage
further depletes the barrier near to the negative electrode,
enhancing its insulating state. In contrast, electrons attracted to
the positive electrode leave behind available donor states and
stimulate the appearance of the metallic state on the positive
side. This simple band bending picture of figure 3, based
on Poisson’s equation, remains remarkably valid for SCEG
heterogeneous systems, even though a simple band structure
description can become questionable. Oka and Nagaosa
recently showed how density matrix renormalization group
calculations can be used to map a modified Poisson’s equation
of the form [1]:

$$\frac{d^2V(x)}{dx^2} = -\frac{e}{\hbar} (n(V(x)) - n_d)$$

where \(n(V(x))\) can be represented in a linearized form with constant compressibility \(k = -\frac{dn}{dV} = \frac{2}{W} \) outside
the Mott gap \(\Delta\), while \(n = 1\) inside [25]. \(W\) is the
bandwidth. The concentration of carriers is described by the
doping ratio \(\delta\), i.e. impurity concentration (\(N_i\), per atomic
concentration), related to bulk charge density \(n_d = 1 - \delta\).
The equation is solved for \(n\) with fixed \(n_d\). We assume p-
doping for the barriers, following our Mott–Schottky analysis
results. The much lower TEM contrast of the Co/Ni metal
island (figure 1) suggests high porosity of the island, that
possibly has a significant amount of impurities (oxides most
likely). We therefore arbitrarily choose a value significantly
different from 1 for \(n\) in the Co/Ni metal island (\(n = 0.8\).
We use the midpoint technique with deferred corrections
numerical method (Maple™) to solve Poisson’s equation with
the boundary conditions set by the applied voltage. The
depletion layer width can be estimated from the full depletion
analysis as

$$\delta_{\text{depletion}} = \sqrt{2\varepsilon(V_f - V_A)}/eN_f^*$$

Taking an impurity concentration of \(N_i = 2 \times 10^{25} \text{ m}^{-3}\), \(\varepsilon = 4\) and the interface barrier \(V_f = \chi + \Delta - V_M = 0.5 \text{ V}\) (a typical value, calculated as the difference of work function in the metal \(V_M\),
electron affinity \(\chi\), and Mott gap \(\Delta\) in SCEG), we estimate
the depletion layer at zero applied potential to be about 3 nm,
i.e. larger than the width of our barrier, making it therefore
insulating even though a significant doping ratio is present in
our structures. Figure 3 illustrates the variation of the potential
and charge density as a function of applied voltage across the
double barrier structure. A short island length (2 nm) was
chosen in order to calculate and draw the numerical results
on a single scale. One should, however, note that identical

\[ \text{Figure 2. Top: } I V \text{ curve of the Ni–NiO–(CoNi)–NiO–Co double barrier nanowire, measured at 1.5 K, showing reversible resistance switching. Bottom: magnetoresistance curves, under a DC bias held at 30 mV, of the two resistance states, with the color coding corresponding to the branches of the top figure.} \]
Figure 3. (a) Flat-band diagram and (b) band diagram under $V_A$ applied voltage. $V_B$ is the metal/semiconductor barrier height, $V_M$ and $V_I$ are the metal work functions of the electrodes and island, $\chi$ is the electron affinity, $\Delta$ is the gap of the band of width $W$. (c) Results of modified Poisson’s equation calculation of the charge density and voltage in the heterostructure. Doping ratio is $\delta = 0.0005$ inside the barriers, and $\delta = 0.2$ in the island $n = 1$ corresponds to insulating state while $n < 1$ corresponds to metallic. Upper part demonstrates how increasing voltage difference $V_A$ diminishes the insulating thickness of the barrier (bars) on the right side and becomes conducting at 100 mV. Lower part is the electron charge density $n(x)$ for several applied voltage values.

considerations and conclusions are drawn if the central island is longer. It is readily seen that the barrier at lower potential keeps the charge density essentially at $n(x) = 1$, and, therefore its insulating property, under applied bias. The asymmetry comes from the interface at higher potential (left side of figure 3), where deviations from $n(x) = 1$ promote a metallic state, corresponding to the nearly constant potential profile under applied bias, reaching the potential barrier between metal and semiconductor. The first order MI transition abruptly changes the potential profile across the device. The interfaces, essentially 3D systems, allow the filling-driven Mott transition to be of first order and to exhibit hysteresis, following the arguments given in [25].

The observed small value of the voltage threshold triggering the resistance switching is an important consequence of the double junction configuration. This finding, of key importance for device applications, can be explained by the asymmetry of the carrier density next to the two opposite electrodes. For a single barrier, the increasing carrier concentration on one side of the barrier that can promote the conducting state of the system is compensated by the depletion of carriers occurring on the other side.
of the barrier. In the double barrier geometry, the island breaks this inversion symmetry, and similarly bends the two barriers densities at the island/barrier interfaces (figure 3(b)). The carrier accumulations at one of the interfaces can add to the accumulations at one of the electrodes, therefore diminishing the insulator/metal switching voltage. We believe that the accumulations at one of the interfaces can add in mind the different mechanism) [5]. A simple electrostatic model, using a modified Poisson’s equation, provides the quantitative tool explaining how a band bending picture explains our experimental findings. Although we applied this model to strongly correlated systems, other materials with filling-induced metal–insulator transition can be described similarly, in particular when invoking mechanisms of electromigration of vacancies with resulting local modifications of the oxygen content near interfaces [8].

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