Electrically controllable magnetoresistance switching in multifunctional organic based spin-valve devices

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Abstract
In this work a multifunctional organic spintronic device is demonstrated using Tris(8-hydroxyquinolinato)aluminium (Alq3) based vertical spin valves with manganite and cobalt electrodes. The device showed a non-volatile electrical switching with dramatic effects on the spin transport behavior. The multifunctionality is illustrated together with a phenomenological model which explains the interplay between the electrical and magnetic bistability.

Keywords: Spintronics; organic electronics; multifunctional; memory

1. Introduction

Nowadays there is a profusion of examples demonstrating the versatility that organic materials can bring into electronic applications. Within these applications, spintronics is one of the latest to experience the introduction of organic semiconductors. Organic spintronics has already moved its first steps towards important results, spanning from the demonstration of spin injection [1,2] to the achievement of high values of spin-valves magnetoresistance [3,4].

In this work we combine the new field of organic spintronics with one of the long known property of organic semiconductors: the resistive memory capability, extending the potentiality offered by organics electronics. Such device can mimic the behavior of a Spin-MOSFET, one of the desiderata in the last version of International Technology Roadmap to Semiconductor (ITRS 2009).

The devices used are organic-based spin-valves, with LSMO and Co electrodes, with Alq3 as organic transport layer.

2. Experimental

The bottom electrode of the devices was a $1 \times 5$ mm$^2$ strip LSMO 20 nm thick. This film is grown on a matching SrTiO$_3$ $5 \times 10$ mm$^2$ substrate in a Channel Spark Ablation (CSA) machine in a $4 \times 10^{-2}$ mbar oxygen atmosphere, with the substrate kept at 880 °C. The sample was then exposed to air and moved to the organic semiconductor (OS) deposition chamber (base pressure $2 \times 10^{-3}$ mbar). Prior to OS deposition, the LSMO sample was heated to 250 °C for 20 min to recover its surface properties. Alq3 layer was evaporated on top of it at a rate of 0.05 Å/s at room temperature with thicknesses ranging from 120 to 250 nm. Subsequently the sample was exposed to air and brought back to the CSA machine were a 2 nm thick AIOX tunnel barrier was deposited at a $2.5 \times 10^{-2}$ Oxygen pressure. Finally the sample was exposed to air and moved to the metals deposition chamber, were a 40 nm thick Co film was evaporated.
with an electron gun at a $5 \times 10^{-8}$ mbar base pressure. The measurements were carried out in a 4-points cross bar configuration by using a Keithley 236 SMU. The resistances of the LSMO electrodes were few kΩ and that of the Co one was hundreds of Ω for all devices, therefore we exclude any geometrical effects due to the finite size of the device, as well as any contribution of the MR of the electrodes to the device MR [5]. The bias is applied at the LSMO electrode, while the Co is kept at ground.

3. Results and discussion

A strong spin valve effect of about 22% [4], with very sharp and well defined resistance switches was detected at 100 K. To the best of our knowledge this is the highest MR reported so far in organic spintronic devices at this temperature: together with the sharp vertical switches, it indicates the high quality of the devices reported here and of their interfaces. The samples showed an electrical bistability, as clearly visible in the Fig. 1. I-V curves show an hysteresis behaviour. Starting from zero and increasing the bias it is possible to reach a threshold voltage ($V_{th}^+$) at which the current suddenly increase and the device switches to a lower resistance state. Going back to zero and applying a negative bias, we can bring back the device to a higher resistance state by reaching the negative voltage threshold $V_{th}^-$. In this device the resistance goes from 362 kOhm to 4.6 MOhm.

The interesting part is that the change of resistance has dramatic effect on the magnetic bistability, i.e. the Spin-Valve Magnetoresistance (SVMR). As clearly visible from Fig. 2, in the low resistance state we have the normal spin-valve effect, while, after switching the device in the high resistance state, no more SVMR is visible. It is worth nothing, that all the MR measures are taken at the bias of $-100$ mV (electrons are injected from the LSMO electrode), after the application of a “programming” voltage higher than $V_{th}^+$ or $V_{th}^-$. 

It is interesting also that the spin-valve can be set in an intermediate state, with an intermediate resistance and a lower SVMR.

In order to explain such multifunctional effect we expanded a phenomenological model developed by Rozenberg et al. [6], adding the spin to the charge transport and obtaining the combined effect we observed in ours samples. Such model is based on the reduction of hopping site due to charge trapping at the interface with one of the electrodes. Lower number of hopping sites correspond to lower charge mobility which in turn raises the charge transit time through

![Fig. 1. I-V curve taken at 100 K, showing the hysteretic behaviour typical of bipolar resistive switching.](image1)

![Fig. 2. On the left schematics of the model are shown, on the right the corresponding Magnetoresistance of the device.](image2)
the organic layer. If this time is higher than the spin coherence life time no more SVMR is visible. A Monte Carlo simulation as been performed in order to verify such hypothesis.

We speculate that the observed control of magnetoresistance can lead to attractive solutions for some standing issues in magnetic storage and more generally in spintronic processing. For example, in a cross bar geometry, the devices can be all set to high resistance state, while setting to low resistance state only the one we want to read. This can reduce the sneak paths issue typical of passive array.

The effect can be also used as an on-off switch of the spin polarization of current flowing along some circuit line.

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