## Magnetoresistance and energy model of Alq3-based spintronic devices.

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Spin transport in organic semiconductors has been receiving widespread attention since the first experimental demonstration of magnetoresistive effects (change in resistance under an applied magnetic field) in hybrid ferromagnetic/organic/ferromagnetic structures [1]. Continuous effort in the field has led to the realization, for example, of vertical organic spintronic devices with different organic semiconductor layers [2,3] or organic tunnel barriers [4]. However, there is still a lack of understanding on the mechanism that governs spin injection and transport in organics, leading to general disagreement even on the expected sign of the devices output magnetoresistance.

With the aim to clarify the spin transport behaviour in organic semiconductors, we present new results on hybrid inorganic/organic spin valves with the most successful up-to-date combination of materials [2-6]. The highly spin polarized manganite La2/3Sr1/3MnO3 and Cobalt have been used as ferromagnetic electrodes for spin injection into thick layers (up to 200 nm) of tris(8-hydroxyquinoline)aluminum(III) (Alq3). In a critical design improvement, we have for the first time introduced an artificial tunnel barrier (Al2O3 or LiF) between the organic and the Co top electrode to study its influence on spin injection into organic semiconductors and to improve the chemical stability and reproducibility of the devices.

In our manuscript we: explore the importance of artificial tunnel barriers for spin injection in organics, record room temperature magnetoresistance, demonstrate that only ferromagnetic electrodes and not organic semiconductor limit device output and, finally, sketch an energy diagram able to explain negative magnetoresistance in LSMO/Alq3/Co spin valves.

Our work is a new step forward in organic spintronics, as we prove that organic semiconductors do not have a clear limit for room temperature performance with the adequate ferromagnets, and we present a reliable model that could be easily extrapolated to predict the output of different materials combinations in hybrid spin valves.

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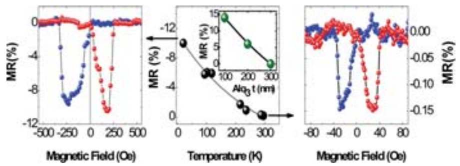


**Figure 1. Magnetoresistance of a La0.7Sr0.3MnO3/Alq3(100 nm)/Al2O3/Co spin valve. a, Inverse spin valve effect at 20 K, showing a maximum value of 11%. b, magnetoresistance values with temperature, showing the decrease, but the persistence of the effect up to room temperature. c, room temperature inverse spin valve effect. The magnetoresistance of each individual electrode was carefully studied, enabling us to rule out anisotropic MR as the origin of our findings. A small background non-hysteretic signal, probably intrinsic to the organic semiconductor layer, was subtracted in every case to clearly show the hysteretic spin valve effect.**

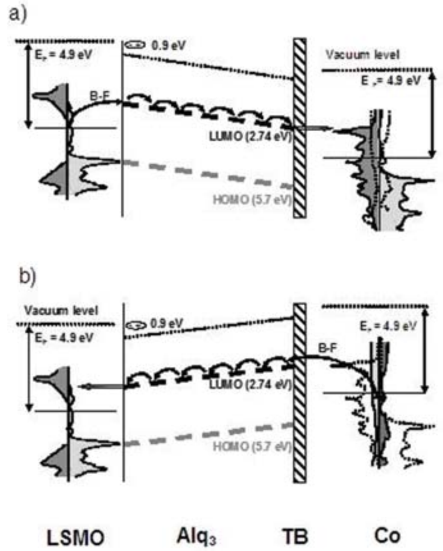


**Figure 2. Energy diagram for a La0.7Sr0.3MnO3/Alq3/Tunnel Barrier/Co organic spin valve. a, Case when carriers are flowing from the LSMO to the d-states of Cobalt (continuous line, coloured plot). Cobalt s-states are also indicated by a dashed line b, Case when carriers are flowing from the s-states of Co (continuous line) to the LSMO. For the Cobalt DOS, d-states are also indicated by a dashed line. Light (dark) grey plots indicated spin up (down) polarization, respectively. Arrows indicate the carrier paths, including the resonance effect across the interfaces. B-F account for the multistep injection process proposed by Baldo and Forrest**