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On the origin of decay of spin current with temperature in organic spintronic devices

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ABSTRACT

This article addresses the most challenging question facing the organic spintronics community today – what causes the universal loss of Giant Magnetoresistance (GMR) signal in organic spin valve devices made with different spin-polarized electrodes and organic semiconductor spacers? Careful analysis of our own and other experimental results available in literature indicate that transition of transport from polaron tunneling limit (suggested by the variable range hopping model) to thermally activated hopping limit (in the temperature range of 40–58 K) marks the most significant decrease of spin relaxation in organic semiconductors. With increasing occupancy of the available hopping sites by the thermally activated carriers, chances of spin flip inside the organic semiconductors increases significantly causing fast spin relaxation in the spin-valves.

1. Introduction

Organic spintronic components like spin valves (SV) and magnetic tunnel junctions (MTJ) have been extensively studied over the last few years [1–8]. The interest primarily lies in the anticipated large spin diffusion length in organic semiconductors (OS) – both pi-conjugated polymers and small molecules, due to their low spin–orbit and hyperfine coupling strength [3]. Although significant GMR response has been observed in many organic SV devices at low temperatures, the response dies out drastically with increasing temperature, leaving no [1] or very small [2,4] spin response at room temperature. This remains the biggest challenge for organic spintronics, rendering them unattractive for applications.

SVs made with different spin injecting and detecting electrodes as well as OS spacers have been reported in literature [1,5,6], among which half-metallic $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) based SVs make up a significant number [3,5]. One of the primary explanations for loss of spin signal in these organic SVs is the loss of spin polarization in LSMO electrode [9]. This is argued despite of the fact that SVs made from transition metal electrodes (Fe, Ni) with higher spin polarization at room temperature have also failed to show improvement [7,10]. Therefore, the question as to what are the decisive factors responsible for disappearance of spin response in OS based spintronic components, still remain unanswered.

In the present communication, we report results from LSMO/regio-regular poly (3-hexyl thiophene) (RRP3HT)/Co based SV devices together with the bare LSMO films. Our results show that loss in spin polarization in LSMO surface, and hence the LSMO/RRP3HT interface is not the only factor destroying the spin response in the SVs and that spin information is also lost in the OS layer. RRP3HT is chosen as the spacer material due to its widespread usage in organic electronics and the background information available in literature. The mobility of RRP3HT reported in field effect transistor (FET) geometry is 0.1 $cm^2 V^{-1} s^{-1}$ [11] while that in the vertical structure is approximately

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$2 \times 10^{-5}\,cm^2\,V^{-1}\,s^{-1}$ [12]. This high mobility value is among the highest in its class due to self-organization of polymer chains forming a lamellae structure and delocalization of the charge carriers inside the lamellae. This high mobility ideally suggests better transport of spin polarized carriers without much loss in their spin information [5] as the carrier will spend less time in the spacer and can reach the counter electrode before their spin relaxation time. However, our experimental results show that better mobility in RRP3HT alone is unable to improve efficiency of spin-polarized (SP) transport significantly. We suggest that with increasing temperature, the numbers of thermally activated charge carriers are increased inside the OS and this increased carrier concentration eventually hinders SP transport causing an universal loss of GMR signal in all OS based SV devices.

2. Experimental methods

First LSMO films were grown on $SrTiO_3$ (100) substrates using pulsed laser deposition at a substrate temperature of 700 °C and oxygen pressure of 0.25 Torr followed by post-annealing treatments in flowing oxygen. After cleaning the LSMO films with acetone and isopropyl alcohol, RRP3HT films were spin-coated on top of LSMO from 10 mg/ml chloroform solutions and annealed at 80 °C for 10 h in a nitrogen-filled glove-box. Finally 10 nm of Co film and an Al capping layer was vacuum-evaporated on top of the RRP3HT films without breaking the vacuum and using the same mask to prevent oxidation of the Co film. The devices have a typical area of 1.3 mm^2. The RRP3HT film thickness measured with AFM before the Co deposition was ~100 nm while the effective thickness is found to be 70–80 nm from charge extraction by linearly increasing voltage (CELIV) measurements. In CELIV, capacitance between two metallic electrodes is measured and from the known dielectric constant of the media between the two electrodes, distance between the electrodes is calculated. So, the measured thickness of RRP3HT indicates 20–30 nm penetration of Co into the soft polymer film during evaporation. The MR of the SVs was measured using four probes, one pair of current and voltage leads connected to the LSMO while the other pair to the Al, varying the in-plane magnetic field between +300 mT and −300 mT and in the temperature range 5–300 K. In all cases, current was kept constant and the change in voltage was measured.

3. Results and discussion

Fig. 1 shows SV response of a typical LSMO/RRP3HT/Co spin valve structure (shown in the inset) at 5 K. The GMR response, calculated using the equation $(R_{AP} - R_P)/R_P$ (where R_{AP} and R_P are the resistances in the antiparallel and parallel state of the device, respectively), is highest at 5 K suggesting efficient spin injection and detection with LSMO and Co electrodes, respectively, and efficient spin transport through the spacer RRP3HT layer. Highest GMR response of more than 100% was observed with 20 nA current, i.e. ≈2.15 V (Fig. 1).

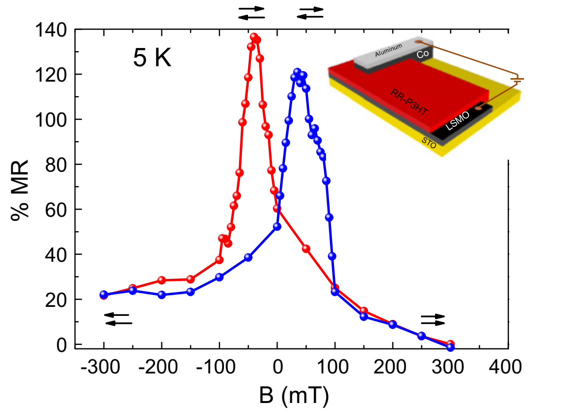

Fig. 1. GMR response of a typical LSMO/RRP3HT/Co spin valve at 5 K measured with 20 nA current. Arrows showing parallel and anti-parallel orientations of the device for upward and downward field sweeps. Inset shows schematic diagram of a spin valve device.

However, with increasing temperature above 20 K, the GMR response starts to decrease almost monotonically and nearly disappears around room temperature (Fig. 2). GMR vs. temperature plot has been shown and discussed in more detail later. Similar temperature dependence of GMR response of LSMO based organic SVs have also been reported by other groups [4,9] and were mainly explained by the loss of spin polarization at the LSMO film surface or LSMO/OS interface. We have previously observed that magnetization, hence spin-polarization, of bulk LSMO films remain high until 300 K [13] but, nevertheless, the concerns about loss of spin-polarization of LSMO film surfaces are justified [14]. We, therefore, further investigate the role of LSMO surface spin polarization on the loss of GMR response of the SVs.

We measured the sheet resistance of LSMO film surface in presence of a varying magnetic field between −300 and +300 mT and calculated the low- field MR (*LFMR*) from the measured data using the formula MR = $[R(B) - R_{300}]/R_{300}$, where $R(B)$ is the sheet resistance of the LSMO at the magnetic field B and R_{300} is the sheet resistance at 300 mT field.

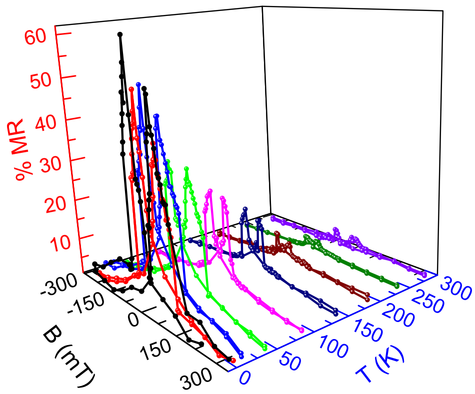

Fig. 2. GMR response of a typical LSMO/RRP3HT/Co spin valve as a function of temperature with 100 nA current.