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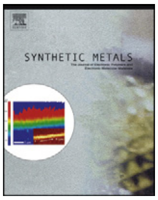

Organic spin-valves based on fullerene C_{60}

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ABSTRACT

Recent work suggests that the spin-transport length in organic semiconductors is limited by hyperfine coupling. Therefore, to potentially overcome this limitation, we fabricated spin-valves based on C_{60} for which the hyperfine coupling is minute. However, our devices do not show a significantly larger spin-diffusion length. This suggests that either a mechanism other than hyperfine coupling causes the loss of spin-polarization, or that in thick devices an increasing conductivity mismatch limits spin-injection.

1. Introduction

Spintronics is a scientific and engineering discipline that incorporates additional functionality based on controlling and manipulating the electron spin degree of freedom into electronic devices. The spin is manipulated through a variety of spin-dependent phenomena. The spin-valve is the principal spintronic device and consists, in its most basic form, of two ferromagnetic electrodes separated by a non-magnetic spacer layer. The two ferromagnetic electrodes exhibit two distinct switching fields, and the device can be switched between a parallel and antiparallel magnetization configuration using an applied magnetic field, H (in this paper, we will use $B = \mu_0 H$, where μ_0 is the magnetic constant, to specify the strength of the applied magnetic field). The difference in resistance between the two configurations is referred to as Giant magnetoresistance (GMR) [1,2] and has a typical magnitude of 10% or more. In GMR devices, typically metallic spacer layers are used, and the spin-polarized current is injected into and transported through the spacer layer. If a thin layer of insulator is chosen as the spacer-layer instead of a metal, the corresponding effect is called tunneling magnetoresistance (TMR) [3,4], because the spin-polarized carriers tunnel through the insulating layer. Using semiconductors as spacer-layers would enable the design of spintronic logic devices, analogous to transistor logic in ordinary charge-based electronics. However, spin-injection into semiconductors continues to be challenging, in part because of the conductivity mismatch problem [5]. The search for new materials systems is therefore ongoing.

Recently, there has been increasing interest in using organic semiconductors for spintronics, partly because of their long spin relaxation times [6,7]. The first demonstration of an organic spintronic device employed a planar structure of $La_{2/3}Sr_{1/3}MnO_3$ (LSMO) electrodes separated by an ≈100 nm long channel of α-sexithiophene [8]. The first vertical organic spin valve device, which utilized LSMO and Co as the ferromagnetic layers and tris-(8-hydroxyquinoline) aluminum (Alq_3) as the organic semiconductor spacer, was demonstrated by Xiong et al. [9] and exhibited a clear spin-valve effect.

The quality of spintronic devices for memory storage and logic operations is dependent upon the reliability of injecting and maintaining a spin polarized signal. Therefore the spin-injection efficiency and spin-diffusion length are amongst the most important parameters characterizing the performance of a spin-valve.

The spin diffusion length is determined by both the mobility of the carriers and the spin relaxation time. Whereas the long spin relaxation times [6,7] in organics are promising, their weakness is a typically low mobility. Mobilities are often less than 0.1 $cm^2\,(Vs)^{-1}$. Organic thin films consequently have a very large resistance, and therefore the conductivity mismatch problem, mentioned earlier as a principal obstacle to efficient spin-injection into semiconductors, may be particularly severe. For an in-depth description of the conductivity mismatch problem the reader may consult the original publication [5]. Only a short description is given here. When the transport through the device is diffusive, a finite conductivity causes drops of chemical potential across the various layers of the device. The chemical potential is spin-dependent only close to the electrode interface. If the device resistance is dominated

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by the bulk resistance of the semiconductor, most of the chemical potential drop occurs across the semiconducting layer, and this drop is spin-independent. This spin-independent voltage drop drives a spin-independent current, making the detection of magnetoconductance difficult. However, large injection resistance typical of tunneling barriers are expected to mitigate this problem [10]. This possible loophole to the conductivity mismatch problem is particularly relevant for organic spintronics since carrier injection often occurs by tunneling. We note that whereas the conductivity mismatch problem is well established for inorganic semiconductor spintronics, the situation is much less understood in organic spintronics. Nevertheless, significant progress in the modeling of organic spintronics devices has been made (Refs. [11,12,7] and references therein).

Spin relaxation times depend predominantly upon two mechanisms: spin–orbit coupling and hyperfine interaction. Spin–orbit coupling results from a particle's spin coupling to a magnetic field or, if we change reference frames, moving through an electric field. In organic semiconductors, spin–orbit coupling is usually weaker than the hyperfine coupling and therefore not the dominant spin-relaxation mechanism. This was most clearly demonstrated by studies of the organic magnetoresistive effect ([13–16] and references therein). Indeed, a theory for spin-diffusion in disordered organic semiconductor devices based on hyperfine coupling was recently developed [17]. The importance of hyperfine coupling on the spin-diffusion length in organic spin-valves was experimentally verified in a recent work that demonstrated enhanced spin-diffusion lengths in deuterated compounds [18].

We fabricated spin-valves using a C_{60} film as the spacer layer to potentially overcome the limitations imposed by hyperfine coupling. C_{60} has orders of magnitude smaller hyperfine coupling than most other organic materials [19]. Carbon exhibits hyperfine coupling only through the rare ^{13}C isotope. Most other organic materials exhibit strong hyperfine coupling because of the abundance of hydrogen in the molecular structure.

2. Device fabrication and measurements

Our devices were fabricated on $SrTiO_3$ (STO) (0 0 1) substrates. On these substrates 1 mm wide, 60 nm thick LSMO films were fabricated by pulsed laser deposition through a shadow mask. The LSMO deposition was performed with a KrF excimer laser (248 nm) in 320 mTorr O_2 at 700 C with a laser fluence of 1–1.5 J/cm^2, followed by post-growth cooling in 300 Torr O_2. The LSMO covered substrates were washed in several solvents using an ultrasonic cleaner and handled in a class 1000 clean-room. As the organic semiconductor spacer layer, a C_{60} layer was fabricated by thermal evaporation in high vacuum at a rate of 0.1 nm/s. Then a 0.5 mm wide, 15 nm thick Co top electrode covered by 30 nm of Al was deposited by electron beam evaporation at a rate of 0.1 nm/s. All fabrication steps involving the organic layer and the top electrode were performed inside a glove-box or inside the glove-box-integrated vacuum evaporation chamber. The device structure is shown schematically in Fig. 1. The substrates were then mounted inside a closed-cycle He cryostat located between the poles of an electromagnet. Current–voltage (*I*–*V*) and magnetoresistance (MR) measurements were performed using a Keithley 2400 source measure unit, with the positive pole connected to the LSMO electrode. Two or four point measurements were performed depending on the relative size of the junction and electrode resistance. Fig. 1 shows the experimental setup for a four point measurement. The molecular structure of C_{60} is shown in Fig. 2 together with the energetic location of its highest occupied and lowest unoccupied molecular orbitals, HOMO and LUMO, respectively. For comparison, the workfunctions of LSMO and Co are also shown in [9].

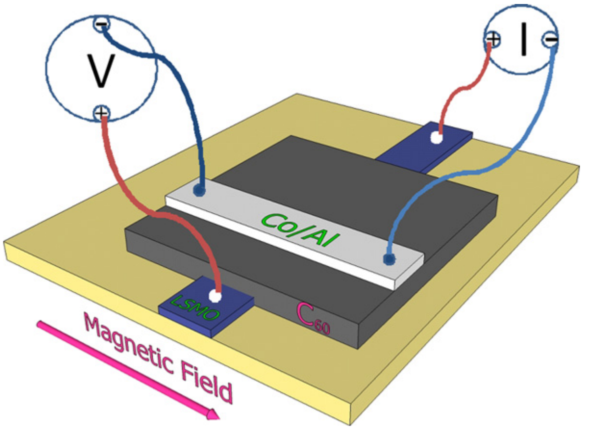

Fig. 1. Structure of LSMO/C_{60}/Co spin-valve and diagram of the 4-point measurement.

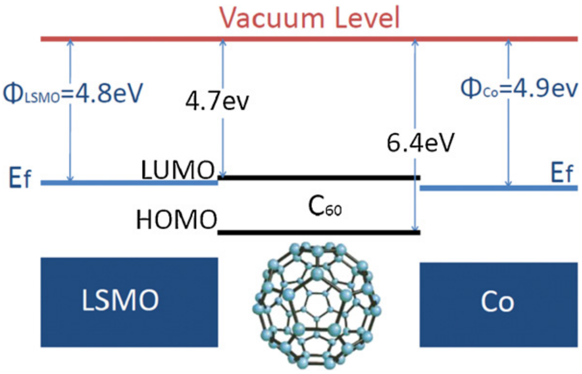

Fig. 2. Schematic band diagram of the LSMO/C_{60}/Co spin-valve showing the Fermi levels and the work functions of the two FM electrodes, LSMO and Co, respectively, and the HOMO and LUMO levels of C_{60}. The diagram is meant to be qualitative, as it neglects any interface dipoles and other effects that modify the band diagram in real devices. The inset shows the chemical structure of C_{60}.

3. Experimental results

Fig. 3, inset, shows a magnetoresistance trace, $\Delta R/R$, measured in a LSMO/C_{60} (40 nm)/Co spin-valve at 15 K. The figure shows that

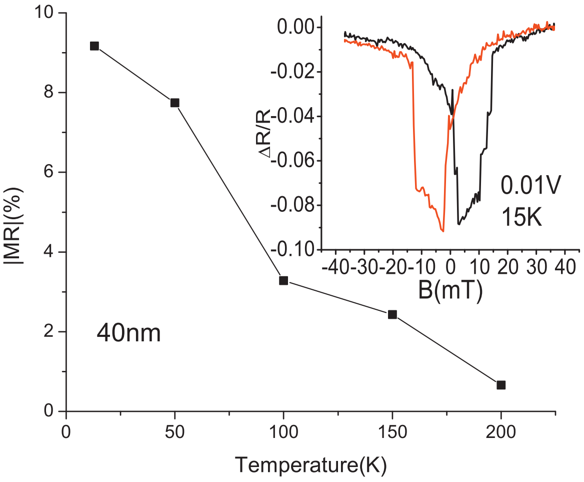

Fig. 3. Temperature dependence of the magnetoresistance, $\Delta R/R$, measured in a LSMO/C_{60} (40 nm)/Co spin-valve. The inset shows the $\Delta R/R$ at 15 K.