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Planar organic spin valves using nanostructured $Ni_{80}Fe_{20}$ magnetic contacts

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ABSTRACT

Planar organic spin valves were fabricated by evaporating organic semiconductor PTCDI-C_{13} onto pairs of patterned $Ni_{80}Fe_{20}$ magnetic nanowires separated by 120 nm. Control over the relative alignment of magnetisation in the nanowires was achieved by including a domain wall 'nucleation pad' at the end of one of the wires to ensure a large separation in magnetic switching fields. Switching behaviour was investigated by optical and X-ray magnetic imaging. Room temperature organic magnetoresistance of −0.35% was observed, which is large compared to that achieved in vertical spin valves with similar materials. We attribute the enhanced performance of the planar geometry to the deposition of the semiconductor on top of the metal, which improves the quality of metal–semiconductor interfaces compared to the metal-on-semiconductor interfaces in vertical spin valves.

1. Introduction

Organic semiconductors (OSCs) have attracted interest as potential charge/spin transport materials for use in spintronic devices, in which electron spin as well as charge is used to perform operations. Despite their low charge carrier mobilities, OSCs are in demand due to their low spin–orbit coupling and weak hyperfine interactions, which help to maintain spin polarisation for longer distances than is achievable in inorganic materials [1]. The scope and rationale for introducing spin transport phenomena into OSC devices (and *vice versa*) was reviewed by Dediu et al. [2].

The most significant component in spintronic technology to date is the spin valve. This consists of two ferromagnetic (FM) contacts connected by a non-magnetic layer, which couples magnetic behaviour to electrical transport. When current flows through the device, one FM contact acts to polarise and the other to analyse the spin of the charge carriers, such that the resistance of the device depends on the relative orientation of the FM contact magnetisations. The magnetoresistance, MR, of the device is defined as

$$MR(\%) = \frac{R_{AP} - R_P}{R_P} \times 100\% \quad (1)$$

where R_P and R_{AP} are the resistance values of the device when the FM contact magnetisations are parallel (P) and anti-parallel (AP), respectively. The magnitude of the MR depends on the spin polarisation in the FM contacts, the physical mechanism responsible for the resistance (tunnelling or scattering at layer interfaces) and the material used in the non-magnetic layer [3]. When the non-magnetic layer is an organic semiconductor, *negative* MR is commonly observed ($R_{AP} < R_P$) [4–8]. A possible explanation

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has been proposed by Schulz et al. [7], based on the assumption that injected carriers do not reflect the bulk (average) spin polarisation of the contact, but the spin polarisation at the energy level corresponding to the relevant OSC transport level (highest occupied molecular orbital [HOMO] for hole transport; lowest unoccupied molecular orbital [LUMO] for electron transport), which may be negative compared to bulk polarisation.

In principle, the use of an OSC as the non-magnetic layer in the spin valve should enhance the MR, due to their minimal interaction with charge carrier spin. Indeed, organic spin valves (OSVs) at cryogenic temperatures have been observed with MR values between −40% and −300% [8–10]. However, at room temperature, the MR commonly drops to around −0.1% [4–7]. Such low room temperature MR values could be due to the vertical geometry (layers stacked on top of each other) employed to achieve sub-100 nm contact spacing, since the topmost interface suffers damage during the deposition of the FM contact onto the inherently soft OSC layer. Sun et al. demonstrated the importance of the organic/FM interface quality by showing that OSVs protected by a frozen xenon buffer layer during deposition of the top FM contact exhibited a 17-fold increase in MR compared to unprotected OSVs [9], but at the expense of very high overall resistance, and a cryogenic manufacturing step.

The top contact problem can be avoided altogether by first preparing in-plane (rather than vertical) FM contact substrates, and deposit OSC over them to form bottom contacts only. However, the preparation of suitable substrates poses considerable challenges: Contact spacing ('channel length', L) in the order 100 nm or less, channel width $W \gg L$, and separate coercivities to allow both P, and AP, configurations. While the first two demands suggest etching a channel into a film of a given FM metal, the latter suggests the use of different FM metals. We here resolve this issue by using electron beam lithography (EBL) on a magnetically soft (low coercivity) FM film to manufacture low L/high W channels, and engineer separate coercivities in the same FM metal by shaping the contacts differently. A similar planar spin-valve has previously been demonstrated using graphene [11] but the MR was extremely small. The planar OSV reported by Kawasugi et al. [12] had a much longer gap between FM contacts, deposited both contacts on top of the organic (rather organic on top of both contacts), and showed no measurable MR.

2. Experimental

Planar OSVs were fabricated in three stages. First, five pairs of magnetic contact nanowires (Fig. 1) were patterned by electron beam lithography into a poly(methylmethacrylate) (PMMA) resist, thermal evaporation of Permalloy ($Ni_{80}Fe_{20}$) and subsequent lift-off of the remaining resist. Lithography was performed at an electron beam energy of 10 keV, designing the nanowire spacing to compensate for the narrowing of the channel between them due to the dose proximity effect (additional removal of resist caused by the overlap of neighboring electron beam dose profiles). Second, non-magnetic Ti(20 nm)/

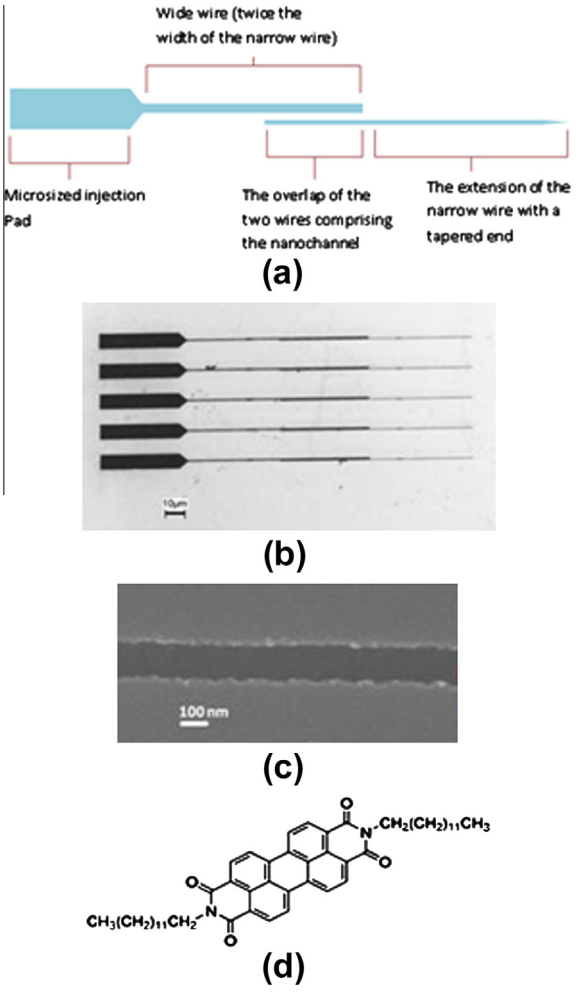

Fig. 1. (a) Schematic design of contact wire pair. Note the different width of the nanowires, and the tapered end of the thinner wire. (b) Electron micrograph of a group of five contact wire pairs. (c) A higher resolution image of the channel region of one contact wire pair (Magnetic wires appear light and the channel dark). (d) Structure of the PTCDI-C_{13} organic semiconductor used here.

Au(200 nm) electrodes were fabricated on top of the nanowires using photolithography, contacting the five pairs of nanowires in parallel, to enable electrical connectivity using spring clips. The non-magnetic electrodes were separated by 150 μm to ensure that current flowed through the magnetic nanowires. These structures were comprehensively characterised for their magnetic behaviour first, before completing the planar OSVs by evaporating a 50 nm thick film of the low molecular weight, electron-transporting OSC N,N′-ditridecyl perylene diimide with α,ω–$C_{13}H_{27}$ alkyl chains (PTCDI-C_{13}; Fig. 1d). We preferred PTCDI-C_{13} over Alq_3, which was often used in OSV research previously [7–10]. The prevalence of Alq_3 is probably for historic [13] rather than scientific reasons: PTCDI-C_{13} has higher charge carrier mobility, a deeper LUMO for better electron injection [14,15] and avoids a metal core that may act as spin scattering centre.

Two sets of samples were prepared: one onto intrinsically doped (high resistivity) Si (001) with a native oxide