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# Organic spintronics: The case of Fe/$Alq_3$/Co spin-valve devices

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**Abstract**

We have embarked on studying magneto-transport response of organic spin-valves made of evaporated $Alq_3$ spacer sandwiched between two ferromagnetic (FM) electrodes with spin-injecting capability. Recently, we have fabricated and completed studies on organic spin-valve devices using half-metallic manganites as one of the spin-injecting FM electrode, which have shown giant magnetoresistance (GMR) of up to 40% at 11 K. Also we found that the GMR response decreases at higher temperatures, and actually disappears at temperatures above ∼180 K, partially because the FM manganite loses its magnetic properties at ambient temperature. In order to realize room temperature organic spin-valve devices, we have begun studying spin-valve devices where both spin-injecting FM electrodes have high Curie temperatures, and thus maintain their magnetic properties at ambient temperature. In this paper we report our preliminary investigations of Fe/$Alq_3$/Co spin-valves, where both electrodes are regular, d-band metallic FM's. We found that these devices show GMR with maximum of about 5% at 11 K. However, at elevated temperatures the GMR value steeply decreases, and in fact vanishes at 90 K. We attribute this decrease to the increase of the spin-lattice relaxation rate of the injected spin-1/2 carriers in the $Alq_3$ spacer at elevated temperatures, where there is also change in the spin carrier injection mechanism at the Fe/$Alq_3$ interface.


*Keywords:* Organic spintronics; Spin-valve devices; Magnetoresistance; Spin-injection; Spin-polarized transport

## 1. Introduction

Spintronics, or spin-electronics [1,2] is a relatively new research field where in addition to the electron charge transport electron spin transport is used. This has the potential to revolutionalize applications such as magnetic recording and magnetic memory [3,4]. The spin-polarized injection, transport, and detection can be readily studied using spin-valve devices. A spin-valve is a layered structure of magnetic and non-magnetic (spacer) materials of which electrical resistance depends on the spin state of electrons that pass through the device. The discovery of giant magnetoresistance (GMR) in metallic spin-valves [5] has further advanced the field of spintronics, and potential applications have naturally followed. Intense research efforts have been recently devoted to extending the spin-dependent transport effects to semiconductors sandwiched between the two spin-injecting ferromagnetic (FM) electrodes [6–8]; however, spin-valves with inorganic semiconductors have not as yet been achieved. Due to their inherent lattice flexibility and long spin coherence [9], π-conjugated organic semiconductors (OSEC) may offer a promising alternative to incorporate semiconductors in spin-related devices, with novel added functionalities such as light emission capability.

Recently there has been substantial research progress in the field of organic spintronics, which include horizontal and vertical spin-valves [10,11], spin-related organic light emitting diodes (OLED) [12], and spin diodes. We have reported the fabrication of $Alq_3$-based spin-valves where one of the FM electrodes was a half-metallic manganite, for which a maximum GMR value of 40% was measured at 11 K [11]. However, because the manganite ferromagnetic properties weaken at ambient temperature it is desirable to fabricate spin-valve devices where more regular FM's are used as the spin-injecting electrodes. Here, we report the fabrication and studies of Fe/$Alq_3$/Co organic spin-valves, where both spin-injecting electrodes are d-band FM's with high Curie temperatures. With these devices, we obtained GMR response with maximum of 5% at 11 K. We also found that the GMR response decreases at elevated temperatures, and actually vanishes at 90 K. We attribute the GMR steep decrease with temperature to the increase of the spin-lattice relaxation rate

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of the injected spin 1/2 carriers in the $Alq_3$ spacer, where also a change in the spin-injection mechanism across the Fe/$Alq_3$ interface occurs.

## 2. Experimental

The spin-valve devices that we have fabricated are in the form of a vertical sandwiched structure, which consists of two FM electrodes and an evaporated layer of $Alq_3$ OSEC. The electrical current through our simple two-terminal devices was perpendicular to the deposited films, and the direction of the externally applied magnetic field, $H$ was parallel to the films. For the spin-valve devices described here we have chosen iron (Fe) as the bottom FM electrode and cobalt (Co) as the top FM electrode. For FM materials, there is an inequality of the spin population at the Fermi level. This inequality can produce a net spin polarization in the current that transport through the FM material. A FM metal can be used as a source of spin-polarized carriers The spin-polarization value, $P$ is defined in terms of the density of carriers n that have spin up, and spin down, such that $P = [n(\text{up}) - n(\text{down})]/[n(\text{up}) + n(\text{down})]$. Iron and cobalt are metallic FM having spin-polarization injection capability $P$ of about 40 and −34%, respectively [13,14].

The spin-valve device studied here was fabricated on a silicon substrate. Following a cleaning procedure using acetone, the silicon substrate was introduced into the evaporation chamber with a base pressure of $5 \times 10^{-7}$ torr. Firstly, the bottom iron electrode was thermally evaporated with a film thickness of about 25 nm. Without breaking the vacuum, we then deposited the OSEC layer ($Alq_3$) with thickness, $d = 140$ nm; the OSEC layer completely covered the iron electrode to protect it from oxidation in the air. Then we opened the vacuum chamber and changed the shadow mask for the deposition of the top Co electrode. For protecting the deposited Co layer (3.5 nm thick) from oxidation and water contamination, we covered it with an aluminum (Al) contacting film of about 100 nm thick. The obtained active device area was about 2 mm × 3 mm. We used a thickness monitor to measure the film thickness of the various electrodes and OSEC spacer.

The magneto-resistance (MR) properties of the fabricated devices were measured in a closed-cycle refrigerator where the temperature, $T$ was varied from 11 to 300 K. The MR measurements were done by applying a constant voltage between the electrodes while measuring the current, $I$ as a function of the in-plane external magnetic field, $H$. The magnetization properties of the FM electrodes versus $H$ were measured as a function of temperature using the magneto-optic Kerr effect (MOKE). We found that the coercive fields, $H_c$ of the two FM electrodes are different, and this allowed us to change their relative magnetization directions to be either parallel or anti-parallel to each other when $H$ was changed over the range of the two $H_c$'s.

## 3. Results and discussion

We found that the device $I$–$V$ characteristic at low applied bias voltage, $V$ was nonlinear, where the electrical resistance, $R$ increases as the temperature decreases (Fig. 1). The rather steep temperature dependence and the nonlinear $I$–$V$ behavior indicate that carrier injection at the FM interfaces is dominated by a combination of tunneling and thermionic emission [15]. However, at the lowest measured temperature we expect that carrier injection by thermionic emission is minimum and thus tunneling prevails. We also found that devices with $d < 100$ nm showed a linear $I$–$V$ behavior and lack of electroluminescence at high currents. This has led us to believe that due to the softness off the $Alq_3$ layer, the deposited cobalt layer may penetrate into the OSEC spacer in the form of inclusions. We have concluded [11] that organic diodes with an evaporated Co electrode have an "ill-defined" layer with thickness, $d_0 = 100$ nm that may contain a layer of the OSEC with pinholes and cobalt inclusions.

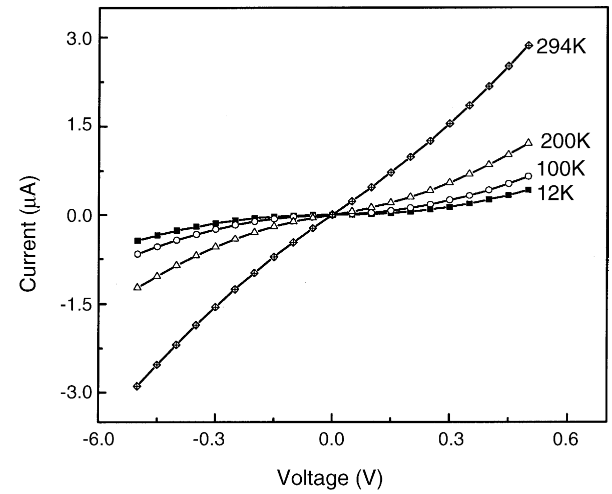


Fig. 1. The $I$–$V$ characteristics of the Fe/$Alq_3$/Co organic spin-valve device.

In order to understand the properties of the charge injection across the FM electrodes, we used the Fowler–Nordheim (FN) type plot of $\ln(I/F^2)$ versus $1/F$, where $F$ is the electric field assumed to be homogeneous ($F = V/d$). Fig. 2 is a FN plot of the obtained device $I$–$V$ curves at different temperatures. We found that the FN curve at 12 K is very different from those at 100 and 200 K. It has been shown [15,16] that at helium temperatures charge injection into the OSEC spacer occurs mainly

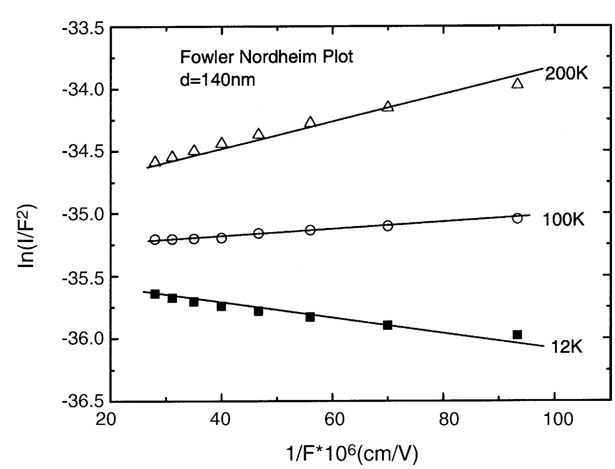


Fig. 2. Fowler–Nordheim plot of the $I$–$V$ curves of Fig. 1.