Spin-Polarised Electron Transport across an Abrupt or Partially Intermixed Fe/GaAs(001) Interface

A. Hirohata^{1,2}, L. R. Fleet^{3,*}, K. Yoshida⁴, H. Kobayashi⁵, Y. Kaneko⁵, Y. Ohno^{5,**}, S. Honda⁶, J. Inoue^{7,**} and H. Ohno⁵

¹ Department of Electronics, University of York, *Heslington, York YO10 5DD, United Kingdom*

² PRESTO, Japan Science and Technology Agency, *4-1-8 Honcho, Kawaguchi 332-0012, Japan*

³ Department of Physics, University of York, *Heslington, York YO10 5DD, United Kingdom*

⁴ Department of Crystalline Materials Science, Nagoya University, *Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan*

⁵ Research Institute of Electrical Communication, Tohoku Univ., *Katahira 3-1-1, Aoba-ku, Sendai 980-8577, Japan*

⁶ College of Engineering Science, University of Tsukuba, *1-1-1 Tennodai, Tsukuba 305-8573, Japan*

⁷ Department of Applied Physics, Nagoya University, *Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan*

There has been significant controversy over the spin-polarised electron transport across Fe/GaAs(001) interfaces, particularly concerning the non-reproducible changes in the spin polarisation. We first overview these previous reports and then provide a solution using abrupt Fe/GaAs(001) interfaces. We demonstrate that an Fe monolayer at the interface controls the spin transport properties and show that abrupt interfaces do not allow sign changes in their injected spin polarisation. The abrupt interface offers reproducible spin transport properties, agreeing with our calculations based on the experimentally observed atomic interface structure. Such junctions are therefore ideal for future spin-transistor applications.

Key words: spin injection, Fe/GaAs(001), spin-polarised transistors, atomic interface structures, epitaxial films

1. Introduction

Si-based metal-oxide-semiconductor field effect transistors (MOSFET) have been following Moore's law since the 1960s, resulting today in approximately 10^8 transistors on a single chip [1]. This fabrication is now based on a 22-nm process rule, as this is the resolution of the optical lithography which uses deep ultra-violet (DUV) light with a wavelength of 193 nm (ArF excimer laser) for patterning. This technique realises a gate length of 18 nm and a high-k dielectric layer thickness of 0.9 nm [2]. It has been predicted, however, that further size reduction to a 16-nm rule will compensate the cost reduction in fabrication, leading to a slow-down in the scaling. According to the International Technology Roadmap for Semiconductors (ITRS) by 2022 extreme ultra-violet (EUV) light with a wavelength of 13.5 nm (soft X-ray) will be introduced. This will allow for the fabrication a MOSFET with a gate length of 4 nm and a dielectric layer thickness of 0.4 nm [1]. This will be the physical limitation for further miniaturisation using such processes. In addition, the rate of increase in the clock speed of transistors is already slowing down, predicted to reach ~100 GHz in 2024. The ITRS also predicts the power consumption in 2022 will be almost 5 times more than current transistors.

Replacing conventional electron charge with spin as the data carrier could greatly improve transistor operation and lead to much higher densities. Accordingly, an all-metallic spin transistor [3] and a spin-polarised field effect transistor [4] have been proposed and subsequently demonstrated. The spin-polarised field effect transistor, proposed by Datta

and Das in 1990, requires efficient spin injection into a semiconductor and highlights the key concepts of three-terminal spintronic devices [3]: Electron spins are injected from a source, modulated by a gate bias and are finally detected at a drain. In an ideal one dimensional spin transistor, the spin orientation is proportional to the gate voltage and so the drain current can be modulated with the spin orientation from a minimum at 180° to a maximum at 360° [5]. As originally proposed by Datta and Das [4], two dimensional electron gas (2DEG) in a semiconductor has been widely exploited as a possible medium for spin-polarised electron transport due to the high in-plane mobility. Successful spin injection from epitaxial Fe into *n*-GaAs(001) has recently been demonstrated at 4 K by Crooker *et al.* [6] who used magneto-optical Kerr imaging to detect the spin polarisation at both the spin injector and detector lateral devices. Spin polarisation of 32% was measured across the Fe/GaAs Schottky junctions.

These junctions, however, exhibit non-reproducible sign changes of their injected spin polarisation P , dependent upon the applied bias. For instance, one of the devices shows negative P between -100 and 0 mV, while another shows it between 0 and $+10$ mV [6]. Such unpredictable transport behaviour prevents these junctions from being implemented into a spin-polarised transistor, as proposed above. Such behaviour has been reported to be induced by their atomic interface structures, as summarised in Table 1, the origins of which are discussed in Section 3.

In this review, we first summarise previous reports on the atomic structures and transport models at the Fe/GaAs(001) interface. We then offer a solution to

Table 1 List of previous reports on the sign change in the spin polarisation.

Spin polarisation	Junction structures	Interfacial atomic structures	References
$P = +32\%$	Fe/AlGaAs/GaAs Schottky junction	Partially intermixed	[7], [8]
$-P$ for $< \pm 100$ mV	Lateral Fe/GaAs/Fe junction	N/A	[6]
TMR < 0	Fe/GaAs (8 nm)/Fe TMR junction	One ideal and another intermixed	[9]
$-P$ for up to $+300$ mV	Fe/GaAs Schottky junction	N/A	[10]

avoid the sign changes in the spin polarisation with an abrupt Fe/GaAs(001) interface successfully grown for the first time [11]. This structure consists of an interface perfectly separating an Fe layer and an As layer. Here, we discuss the reproducibility of its magneto-transport properties by comparing optical and electrical measurements with calculations based on the atomic interface structure extracted from electron microscopy.

2. Fe/GaAs(001) Interface Structures

Demchenko and Lui have proposed potential models for the Fe/GaAs(001) interface [12]. As shown in Fig. 1, there are four distinctive atomic interface structures. The ideal one is the abrupt interface, which sharply separates the Fe and topmost As layers. Here, the As-terminated GaAs surface can maintain the spin polarisation for injection and detection as discussed in Sec. 4.2. When Fe atoms fall in between the topmost As atoms, the interface becomes partially intermixed. By letting the Fe atoms further down between the Ga atoms below the topmost As layer, the fully intermixed interface is realised. When the mixing occurs in the Fe layer with Ga atoms by high-temperature annealing [13], the interface forms Fe_3Ga , which is known to be a magnetically dead layer and spin-polarisation reversal [14]. This interface, however, is only released with high temperature growth or annealing.

Fig. 1 Schematic diagram of possible atomic structures at the Fe/GaAs(001) interfaces (Fe: open circles, Ga: closed circles and As: grey circles).

Most of the interfaces currently reported have been shown to be partially intermixed [8],[15]. Zega *et al.* employed a (2×4) surface reconstruction before the Fe deposition, followed by post-annealing at 473K for 10 minutes [8]. LeBau *et al.* used a $c(4 \times 4)$ surface

reconstruction before the Fe deposition, followed by post-annealing at 473K for 1 hour [15]. To date, only our group has reported the abrupt interface [11], which permits ideal spin-polarised transport across the interface, using the growth conditions detailed in Section 4. It is therefore crucial to control the Fe growth conditions precisely in addition to the GaAs surface treatment.

3. Origins of Spin Polarisation Reversal

There has been contradictory reports over the spin transport across Fe/GaAs interfaces, *i.e.*, non-reproducible sign changes in the injected spin polarisation. Figure 2 shows two possible reasons for this behaviour, both occurring at the interface: tunnelling either through an interface resonant state (IRS) [16]-[18],[20] or from a semiconductor bound state (BS) [19]. IRSs can exist in one of the spin bands and can accommodate a large number of electrons in the vicinity of the Fermi level. This can lead to reversal of the sign of P if the IRS is formed in the minority spin band. These IRSs can then be activated by tuning the bias voltage across the interface to match the IRSs energy levels. The BS, on the other hand, is formed in the depletion layer in GaAs. The shape of the BS strongly depends on the doping profile and can show very weak bias dependence [19].

Fig. 2 Schematic diagram of interface resonant states and bound states which can lead to a reversal of the injected spin polarisation.

Major models to describe these states are listed in

Table 2 List of previous reports on the origin of the sign change in the spin polarisation.

Origins	Systems	Description	References
IRS	Ferromagnet/Insulator/Ferromagnet	Resonant tunnelling, no bias dependence	[16]
IRS	Fe/MgO/Fe	IRS formed in Fe down-spin band	[17]
IRS	Fe/GaAs (8 ML)/bcc Cu	IRS formed in Fe down-spin band	[18]
BS	Fe/GaAs Schottky junction	BS formed in GaAs, significant for spin extraction	[19]
IRS	Epitaxial Fe/GaAs(001)	IRS formed in GaAs, Strong bias dependence	[20]
IRS	Epitaxial Fe/GaAs(001)	IRS formed in GaAs, Interface dependence	[11]

Table 2. As the experiments have revealed bias dependence, as listed in Table 1, the IRS models appear the most promising to explain the interfacial spin transport. It should be noted that the IRS can be form in either the Fe [15],[16] or GaAs layers [20].

4. Abrupt Fe/GaAs(001) Interfaces

Fe/GaAs(001) junctions were fabricated using ultrahigh vacuum (UHV) molecular beam epitaxy (MBE) [11]. A 12-nm-thick *n*-doped GaAs layer ($1 \times 10^{19} \text{ cm}^{-3}$) was deposited onto a GaAs(001) substrate with buffer layers, consisting of GaAs (200 nm)/[AlAs (3 nm)/GaAs (3 nm)]₁₀/GaAs (200 nm)/*n*-GaAs (2 μm , $2 \times 10^{16} \text{ cm}^{-3}$). The GaAs(001) stacks were then capped by As layers and transferred into a second UHV-MBE chamber. The As cap was removed by heating the substrates at 853K until a (2 $\times$ 4) surface reconstruction was observed by reflection high energy electron diffraction (RHEED). The substrates were then cooled down, typically to RT or -100°C , using liquid nitrogen to deposit a 5-nm-thick Fe layer, capped with 3-nm-thick Au film.

4.1 Structural analysis

The above Fe/GaAs(001) junctions were annealed at 200°C for 10 minutes to form atomically flat interfaces [19]. These junctions were then processed for cross-sectional high-resolution (scanning) transmission electron microscopy (HR-(S)TEM) observation using conventional dimpling and low-angle Ar-ion milling techniques.

High angle annular dark field (HAADF) images were taken using a JEOL JEM-2200FS double-Cs-corrected HR-(S)TEM, proving high quality epitaxial growth. An unfiltered image is shown in Fig. 3, clearly indicating the coexistence of two interfaces: (I) abrupt and (II) partially intermixed [11]. Here, almost 95% of the interfaces are found to be abrupt. The partially intermixed interfaces are typically observed at the edges of GaAs terraces, which may be formed to release the lattice mismatch between Fe and GaAs (1.4%).

From Fig. 3, atomic separations between Fe, Ga and As atoms were extracted, shown in Fig. 4. As can be seen, the interface structures for both cases are almost identical except for the Fe atom located between the As atoms for the partially intermixed structure. The inserted Fe atoms are found to shift towards the above Fe layers by $\sim 10\%$. The spin-resolved density of states

(DOS) was calculated for both structures discussed in Section 4.3.

Fig. 3 Cross-sectional HR-TEM image of Fe/GaAs (001), showing the co-existence of (I) abrupt and (II) partially intermixed interfaces.

Fig. 4 Atomic interface structures: (I) abrupt and (II) partially intermixed, extracted from the HR-TEM image shown in Fig. 3, with the spacings shown in nm.

4.2 Transport measurements

The Fe/GaAs(001) samples were also patterned into three-terminal devices using photolithography together with chemical etching and Ar-ion milling [11]. As schematically shown in Fig. 5, a conventional three-terminal method was used to explore transport across the junctions: a spin-polarised electron current

was generated between two contacts (the circuit on the left-hand side), whilst the voltage between the injector and a third contact was measured (the circuit on the right-hand side).

Scanning magneto-optical Kerr effect (MOKE) microscopy was employed to measure the injected spin polarisation over the surface of the GaAs mesa. Figure 5b shows the bias dependence of the MOKE signals for the positions separated from the spin injector. This figure clearly shows monotonous increase of the Kerr rotation with increasing the bias, indicating no sign change in the spin polarisation dependent upon the bias conditions. It should be noted that, in these lateral Fe/GaAs/Fe devices, when flowing a current between two contacts the Schottky junctions are oppositely biased so majority-spin electrons were injected from one contact whilst being extracted from the other. A positive (negative) spin polarisation is achieved in reversed (forward) bias.

Fig. 5 a Schematic diagram of the scanning MOKE setup. b Bias dependence of the MOKE signals with respect to the distance from the spin injector. c, Bias-dependent Hanle curves obtained at 50K with d the peak amplitudes and corresponding spin lifetimes at 50K.

The spin-polarised electron transport was also explored using all-electrical Hanle measurements, as reported in Ref. [11], confirming the monotonic increase in spin polarisation with increasing bias voltage observed using optical methods. Here, the spin lifetime was estimated to be approximately 15 ns at 10K, shown in Fig. 5d, corresponding to a spin diffusion length of approximately 16 μm at 10K. These values are very similar to those previously reported [6].

4.3 Spin transport modelling

In order to model the spin-polarised electron transport across both the abrupt and partially

intermixed interfaces, *ab initio* calculations were performed. Starting with the atomic separations found experimentally (see Fig. 4) the Fe/GaAs(001) interfaces were allowed to relax in order to find their total energy minimum. Both interfaces are found to be energetically stable, the difference of which is almost negligible [22].

Local DOS, at the interface, were then calculated along the Γ point in the two-dimensional Brillouin zone. As shown in Fig. 6, the topmost As layer of the abrupt interface (I) does not possess a peak in the DOS in the vicinity of the Fermi level. This indicates that there are no interface resonant states formed at the abrupt interface. The partially intermixed structure (II), however, has a significant peak for the down-spins at ~ 0.1 eV above the Fermi level. This peak indicates the presence of an interface state which can introduce a much greater amount of down-spin electrons into GaAs. As discussed in Section 3, such a peak can induce a sign change of the spin polarisation. This can explain the bias-dependent sign change in P as all the experimental evidence for the sign change occurs below a few 100 mV, as listed in Table 1. It should be noted that, according to our calculation, such an IRS cannot be formed in Fe.

Fig. 6 Calculated density of states for the topmost As layer for (I) abrupt and (II) partially intermixed interfaces.

The abrupt interface can therefore maintain the spin polarisation while the partially intermixed interface may offer a sign change in the polarisation, dependent upon the amount of interface mixing. This agrees with the experimental results described in Section 4.3 and guarantees reproducible spin injection across abrupt Fe/GaAs(001) junctions.

5. Summary

We have successfully fabricated abrupt Fe/GaAs(001) junctions for the first time and demonstrated reproducible, monotonous spin polarisation of the injected electrons with increasing bias. This behaviour can be explained by modeling the atomic interface structures determined using HR-(S)TEM. These

junctions can be implemented into future spin-polarised transistors.

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Present Addresses

* Department of Materials, Imperial College London, *South Kensington Campus, London SW7 2AZ, United Kingdom*

** College of Engineering Science, University of Tsukuba, *1-1-1 Tennodai, Tsukuba 305-8573, Japan*

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