

Hybrid magnetic/semiconductor spintronic materials and devices

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Elsevier use only: Received date here; revised date here; accepted date here

Abstract

We report our experimental studies of different kinds of magnetic/semiconductor hybrid materials and devices highly promising for the next generation spintronics. The epitaxial Fe films on three III-V Semiconductor surfaces, $\text{In}_x\text{Ga}_{1-x}\text{As}(100)$, $x=0, 1, 0.2$, show a uniaxial magnetic anisotropy in the ultra thin region. This suggests that both interface bonding and the magnetoelastic effect control the magnetic anisotropy. We demonstrate the epitaxial growth of new hybrid spintronic structures, namely, $\text{Fe}_3\text{O}_4/\text{GaAs}$ and $\text{Fe}_3\text{O}_4/\text{MgO}/\text{GaAs}$, where the magnetic oxide has both high Curie temperature and high spin polarisation. Both the magnetisation loops and magneto-resistance curves of $\text{Fe}_3\text{O}_4/\text{GaAs}$ were found to be dominated by a strong uniaxial magnetic anisotropy. We have also fabricated a novel vertical hybrid spin device, i.e. $\text{Co}(15\text{ML})/\text{GaAs}(50\text{nm}, \text{n-type})/\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}(200\text{nm}, \text{n-type})/\text{FeNi}(30\text{nm})$ and observed for the first time a change of the magneto-resistance up to 12% by direct transport measurements, which demonstrated the large spin-injection and the feasibility to fabricate the spin-transistors capable of operating at room temperatures by using magnetic/semiconductor hybrid materials.

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PACS: 75.70.Cn; 82.23.pt; 73.50-h;

Keywords: Spintronics, magnetic ultrathin films, magneto transport;

I. Introduction

Over the past three decades semiconductor electronics has developed dramatically, but with very little attention devoted to incorporating magnetic materials into integrated electronic devices. However, an exciting new field of electronics, spintronics [1], has attracted great attention since the discovery of a giant magneto-resistance (GMR) in magnetic multilayers. The next generation spintronic-devices, combining the advantages of magnetic materials and semiconductors, are expected to be non-volatile,

versatile, fast and capable of simultaneous data storage and processing, while at the same time, consuming less energy. It has the potential to revolutionise the whole IT industry as did the development of the transistor 50 years ago. One of the major challenges in developing this next generation spintronic devices is the integration of magnetic and semiconductor materials. There are three classes of materials systems currently exploited: a) Dilute magnetic semiconductors (DMS) such as $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ and $\text{Cd}_{1-x}\text{Co}_x\text{Se}$ etc, b) Heusler alloys and half-metallic magnetic oxides, such as NiMnSb ,

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CoMnGa and Fe₃O₄ etc. and c) Ferromagnetic metals and alloys, such as Fe, Co, FeNi etc. The DMS have the great advantage of integrating with conventional semiconductor materials and devices and the challenge is to increase their Curie temperatures for room temperature operations [2]. Half-metallic Heusler alloys and half-metallic magnetic oxides have recently attracted great attention for spintronics as they have high polarisation at the Fermi level and high Curie temperatures which are believed to benefit the injection of spin carriers into the semiconductors [3,4]. The most extensively studied hybrid spintronic materials are the ferromagnetic metal/semiconductor hybrid systems [5-9]. The growth of epitaxial ferromagnetic metal/semiconductor hybrid structures was first demonstrated in Fe/GaAs, and it continues to be of interest as a model system for the epitaxial growth of ferromagnetic metals on semiconductors. The Fe/InAs hybrid structure is also an interesting system [10] and high quality bcc Fe has been grown on InAs(100) surface [10]. In this paper, we will highlight our recent studies of both ferromagnetic metal/semiconductor and magnetic oxide/semiconductor hybrid spintronic structures and novel vertical spin-injection devices using a GaAs/AlGaAs membrane. Our results demonstrated that hybrid magnetic/semiconductor structures with well controlled magnetic properties and high Curie temperatures are one of the most promising systems for the development of the next generation spintronics.

II. Sample growth by Molecular Beam Epitaxy

Samples were grown by MBE at pressure of about $1-2 \times 10^{-10}$ mbar. To reduce the intermixing of Fe with Ga, In, or As at the interface, the samples are now grown at room temperature [8]. All stages of growth were monitored by RHEED measurement. The RHEED patterns of a 7ML thick Fe film grown onto three different substrates, namely GaAs, InAs, and In_{0.2}Ga_{0.8}As are shown in Fig. 1. The images confirm the epitaxial growth of bcc Fe on the three different surfaces with the same epitaxial relationship of Fe(100)<001> || In_xGa_{1-x}As(100)<001>, where $x=0, 1, 0.2$. In order to obtain an epitaxial Fe₃O₄ thin film, an Fe film was grown epitaxially on a GaAs substrate at room temperature. After the Fe growth the sample was oxidised *in-situ* with an O₂ partial pressure of 5×10^{-5} mbar at 500 K.

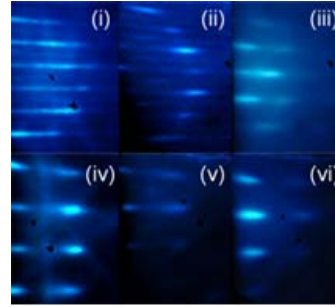


Fig. 1. RHEED images collected before (i, ii, iii) and after (iv, v, vi) deposition of 7ML Fe onto the respective GaAs, InAs and In_{0.2}Ga_{0.8}As surfaces

Fig. 2a shows the RHEED pattern of a 6nm epitaxial Fe₃O₄ thin film on GaAs(100). Further investigation with XPS spectroscopy confirms that the samples only contain magnetite not maghemite, supported by XMCD observation which shows no contribution from the metallic Fe [4]. Fig. 2b shows schematically a proposed alignment of the magnetite cell, which is rotated by 45° relative to the GaAs(100) substrate. The rotation of 45° might be due to the fact that the Fe₃O₄(100)<011> direction, rather than the Fe₃O₄(100)<010>, matches better with GaAs(100)<010> symmetry. The lattice constant of GaAs is 5.654 Å and for Fe₃O₄, it is 8.396 Å. Simple calculation shows the mismatch between Fe₃O₄ and GaAs is about 5.0% if the Fe₃O₄ is rotated by 45° relative to GaAs cell on the (100) plane. We have also grown Fe₃O₄/MgO/GaAs(100) structures

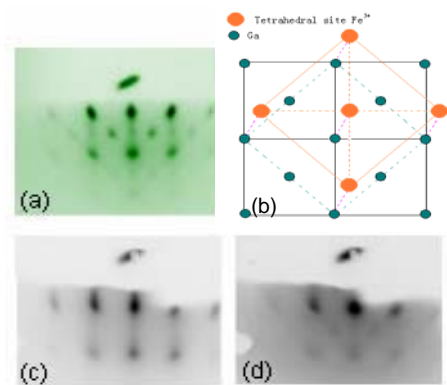


Figure 2. (a) RHEED pattern of Fe₃O₄ (6nm)/GaAs (100), (b) a schematic representation of the lattice positions of Fe₃O₄ on GaAs(100) surface, (c) RHEED pattern of MgO(2nm)/GaAs(100), (d) RHEED pattern of Fe₃O₄(4nm)/MgO(2nm)/GaAs(100). The electron beam was along the [0-11] direction of the GaAs(100) substrate and the beam energy was set at 10.0 kV

incorporating epitaxial MgO as a tunnelling barrier. The MgO, with a lattice constant of 4.213 Å, corresponds with that of the GaAs(100) if it is rotated by 45° with a mismatch of 5.4%. The clear RHEED pattern as included in Fig 2c of a 2nm film shows that epitaxial MgO has been stabilised on GaAs(100). The RHEED pattern suggests that the MgO principle axes appear parallel to GaAs principle axes, despite the relatively large mismatch (25.5%) in this epitaxial relationship. We have further deposited Fe₃O₄ on the MgO layer. As shown by the RHEED image in Fig 2d of a 4nm Fe₃O₄ on the MgO/GaAs, the diffraction pattern is still visible indicating possible epitaxial growth of Fe₃O₄ layer on the MgO buffer layer.

III. Ferromagnetic Metal/III-V Semiconductor systems: Fe/In_xGa_{1-x}As(100)

There are two interesting fundamental issues in ferromagnetic metal (FM)/III-V semiconductor (SC) systems: the first one is the possible interface magnetic dead layer and the second is the uniaxial magnetic anisotropy (UMA) unexpected from the crystal symmetry of bcc Fe. The next generation spintronic devices would require the magnetic material/semiconductor interface to be magnetic in order to achieve highly efficient spin transport. A strong reduction of the magnetization has previously

been found for Fe grown on GaAs [6] which was attributed to the magnetically ‘dead’ layers near the interface. Recently we have used X-ray magnetic circular dichroism (XMCD) to determine the spin and orbital magnetic moments of monolayer and sub-monolayer Fe atoms at the interface [11]. The XMCD technique offers the unique capacity of giving element specific information at sub-monolayer sensitivity. The sample structures were Fe of 1ML and 0.5ML in thickness on a GaAs(100) capped with 7ML of Co and then 2nm of Cr. The spin moments were found to be $m_{\text{spin}}=1.84\pm 0.21\mu_B$ for 0.5ML and $m_{\text{spin}}=1.84\pm 0.11\mu_B$ for 1ML, respectively, comparable to the bulk value of $1.98\mu_B$. These results demonstrate unambiguously that the Fe/GaAs interface is magnetic down to the sub-monolayer region. Kneeder *et al* [7] proposed that the unidirectional nature of Fe-As or Fe-Ga bonds is responsible for the UMA. This might be understood as a “chemical” effect, in which the electronic structure of the Fe atoms near the interface differs distinctly from “normal” bcc Fe. Another picture is that uniaxial magneto-elastic coupling due to anisotropic lattice relaxation is the origin of the uniaxial magnetic anisotropy [12]. In order to determine directly the correlation between the lattice match and the magnetic anisotropy, here we compare directly the Fe films under exactly the same growth conditions on three different structures GaAs, InAs and In_{0.2}Ga_{0.8}As giving mismatches of -1.56%, 5.32% and 0% respectively.

As shown in Fig 3, the MOKE loops taken along the easy (i, iv and v) and hard (ii, iii and vi) axes of the Fe/GaAs, Fe/InAs and Fe/In_{0.2}Ga_{0.8}As samples, respectively, display uniaxial anisotropy in all samples. A rotation of 90° of hard and easy axes in the case of Fe/InAs compared with Fe/GaAs is clearly shown in the figure. This result is in good agreement with previous studies [12] and is thought to be indicative of the influence of the magneto-elastic effect in the establishment of the uniaxial anisotropy. The magneto-elastic effect occurs as a direct result of the compression or expansion of the Fe film in order to fit the GaAs or InAs lattice respectively. Very recently, we have found that the magnetoelastic constants of a 10ML Fe film on GaAs could be 20 times larger than that of bulk Fe,

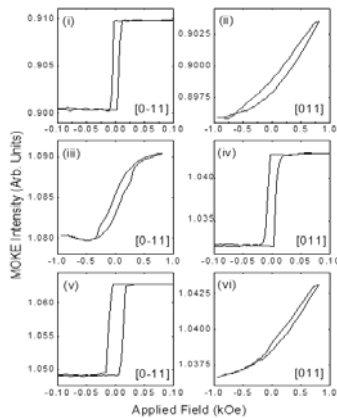


Fig. 3. MOKE loops taken along [0-11] and [011] directions of (i, ii) the Fe/GaAs(100), (iii, iv) Fe/InAs(100) and (v, vi) Fe/In_{0.2}Ga_{0.8}As(100) respectively, show uniaxial anisotropy in all samples. Note the 90° rotation of hard and easy axes in the case of Fe/InAs(100).

showing the importance of magneto-elastic coupling in ultrathin films [13]. However in figure 3 we can also see that the lattice matched system Fe/In_{0.2}Ga_{0.8}As also displays uniaxial characteristics. While this can not be explained by magneto-elasticity induced uniaxial anisotropy the result would seem to support the “chemical” effect. Our results clearly show that both chemical and magneto-elastic effects are in some part responsible for the observed UMA in Fe/III-V semiconductor hybrid structures.

IV. Half metallic/III-V Semiconductor hybrid structure Fe₃O₄/GaAs(100)

The magnetic properties of the Fe₃O₄/GaAs(100) films were studied by *ex-situ* MOKE at room temperature. For a narrow thickness range of about 1.6 nm, S shaped MOKE loops with zero hysteresis were observed indicating that the films are superparamagnetic as observed in the Fe/GaAs films [17]. MOKE observations described magnetic uniaxial anisotropy behaviour over a thickness range of about 2.0 – 6.0 nm as shown in Fig. 4. At a thickness of 2.0 nm, uniaxial magnetic anisotropy is clearly observed, and at about 4.2 nm, the system still shows a uniaxial anisotropy. However, at a thickness of 6.0 nm the cubic anisotropy of the bulk Fe₃O₄ presents and rotates the global easy axis slowly away from the [0-11] direction of the GaAs substrate, which is rotated by 90° with respect to that of the

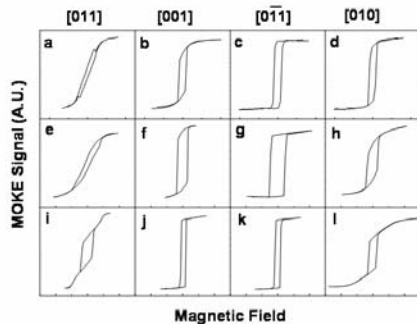


Figure 4. MOKE loops of Fe₃O₄/GaAs(100) of three thicknesses, 2nm (a-d), 4.2nm (e-h), and 6nm (i-l). The directions of the external magnetic field are labelled on top of each column according to the GaAs(100) substrate. Note the magnetic field in the first row (a – d) was between –0.25 and +0.25 KOe, while in other rows (e – l), between –0.75 and +0.75 KOe.

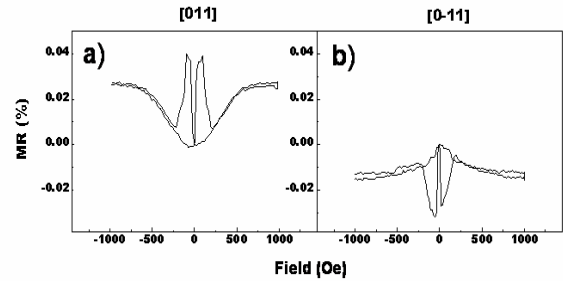


Fig. 5. In plane magnetoresistance curves of Fe₃O₄ (4nm)/GaAs(100) with magnetic field applied along a) the [011] and b) the [0-11] directions.

Au/Fe/GaAs sample of the same thickness and grown at the same time. The saturation field for these samples is observed to be as small as 300Oe as shown in Fig. 4. The small saturation field could be an indication that the samples have reduced anti-phase boundaries (APB).

Fig 5 shows the in plane magneto-resistance (MR) measurements of a 4 nm thick Fe₃O₄/GaAs film with the magnetic field along the easy [0-11] and hard [011] directions. This anisotropic MR response along different directions has not been reported before in Fe₃O₄/MgO [14]. The MR along the hard axes initially decreases when the field is reduced and an increase through an abrupt irreversible jump is observed immediately when the field switches to opposite direction, then it reaches its maximum at the coercive field after which the MR is reduced with another abrupt jump. Then it gradually increases with increasing the field. The opposite has been observed when the measurement is done with the magnetic field aligned along the easy axis. If we ignore the jump at and around the coercive fields, the MR curves would look like ordinary AMR with a negative contribution along the easy direction and positive contribution along the hard direction. This is not an unlikely event as a similar trend has been observed for other magnetite films and has occurred due to the negative spin polarization effect [15]. However, the MR jumps which have been superimposed on the AMR effect might be related to abrupt magnetization change across the possible APB. The change in the resistance could then come from spin dependent domain wall type scattering across the anti-phase boundaries.

V. The Co/GaAs/FeNi Hybrid Spintronics Device

Spin injection and detection from ferromagnetic metals into semiconductors have been achieved by optical techniques [16–18]. However, to integrate with present semiconductor technology direct electrical spin injection and detection are critical. We have recently been studying vertical hybrid devices with the structure, Cr(20ML)/Co(15ML)/GaAs (50nm,n-type) /Al_{0.3}Ga_{0.7}As (200nm,n-type) /FeNi (30nm) [19]. The Co was grown in an UHV chamber and capped with 2nm of Cr. Optical lithography was performed on the thinned sample from the back side to open micron size window. Selective chemical etching was then carried out through window to etches away GaAs and stops at AlGaAs. The surface of the AlGaAs appeared to be very flat when observed under an optical microscope. Once the AlGaAs layer was reached the sample was put into a thermal evaporator and a 30 nm NiFe layer was evaporated and lift-off was performed. The transport measurements were done at room temperature in a current-perpendicular to plan geometry from the NiFe to the Co layer through the AlGaAs/GaAs layer. The field dependent I-V characteristic exhibited a highly rectifying nature and a dependence on the applied field [19]. The most striking feature is a biasing dependent MR characteristic, as shown in Fig. 6. The MR is negligible at low bias current. However, beyond a critical current of 5 μ A the MR increases. A maximum change of about 12% in the MR is observed. This is a much large change compared to ordinary AMR effects of around 1% in FeNi and Co. These direct MR measurements indicate a large room temperature spin injection and detection through the semiconductor layers.

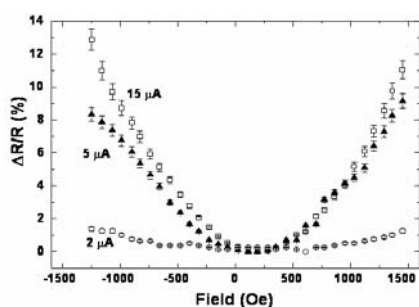


Fig. 6. MR ratio of the vertical spin-device Cr(2nm)/Co(15ML)/GaAs(50nm,n-type) /Al_{0.3}Ga_{0.7}As (200nm, n-type)/FeNi(30nm) at different biasing current.

VI. Summary

We have studied the epitaxial growth, structure, magnetic and magneto transport properties of several hybrid spintronic materials and devices. All the Fe and Fe₃O₄ films in these structures show a strong uniaxial magnetic anisotropy in the ultra thin region due to both the interface bonding and the magnetoelastic effect, which can be controlled for spin-device operations. The new hybrid spintronic structures Fe₃O₄/GaAs and Fe₃O₄/MgO/GaAs with high Curie temperature and high spin polarisation might be one of the most promising systems for high efficient spin-devices. We have also fabricated a novel vertical Co/GaAs/FeNi spintronic device with a GaAs/AlGaAs membrane. This device exhibited rectifying current-voltage (I-V) characteristic, with biasing current dependent MR characteristics. A maximum change of 12% in the MR was observed which demonstrated for the first time the large room temperature spin injection and detection by using direct electronic transport measurements.

Acknowledgement:

The author acknowledges the financial support of the EPSRC, the WR studentship, and WUN network.

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