Magnetic films epitaxially grown on semiconductors

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Abstract

Future ‘magneto-electronics’ exploiting the spin of the electron in addition to its charge will require the injection of spin-polarized electrons from a ferromagnetic metal into a semiconductor. To this purpose an attempt has been made in the present study to avoid the formation of a non-magnetic interface phase or ‘magnetically dead layers’ which have been found in the past in Fe films epitaxially grown on GaAs(0 0 1). Fe(0 0 1) films were grown by molecular beam epitaxy and magnetron sputtering on As depleted GaAs(0 0 1) surfaces held at room temperature. Very good epitaxial growth is achieved by both deposition methods. MBE grown ultrathin films show an enhanced ground state net magnetization and the full bulk magnetic moments at the Fe/GaAs interface in contrast to previous reports. This might be an important step towards semiconductor devices using spin polarized electron transport. The in-plane magnetic anisotropy of the films generally consists of a fourfold and a uniaxial term. At 7 monolayer thickness only a strong uniaxial contribution is observed which is supposed to result from the intrinsic anisotropy of the dangling bonds at the GaAs(0 0 1) surface. This could also be a useful property for future memory or switching applications.

Keywords: Epitaxial films; Magnetic moments; Magnetic anisotropy; Metal–semiconductor interface; Fe on GaAs

1. Introduction

A large potential of technical applications is attributed to future ‘magneto-electronic’ devices, i.e. such devices which in addition to the charge of the electron make use of its spin. As an example a spin-polarized field effect transistor was proposed by Datta and Das [1] some years ago which is based on the injection of spin-polarized electrons from a ferromagnetic source electrode into the semiconductor. The corresponding future technology will most probably require the monolithic integration of ferromagnetic thin film elements into semiconductor circuits. Epitaxial growth of Fe on GaAs, e.g., has been demonstrated in the past [2]. However, ‘magnetically dead layers’ have been found in Fe films grown on GaAs due to the formation of non-ferromagnetic intermetallic compounds at the interfaces [2–4].

From such an interface the injection of highly spin-polarized electrons into the semiconductor
will not be possible. For spin-polarized electron transport across the interface a high magnetic polarization, i.e. large ferromagnetically ordered moments even in the first metallic monolayer at the interface are a necessary condition.

In the past, attempts have been made to overcome this problem, e.g. by passivating the GaAs surface with sulfur [5]. However, in addition to the fact that quantitative information on the magnetic moments of Fe atoms at the interface is lacking it is not clear to what extent the deposition of sulfur changes the electronic states at the interface. In the present study a different approach was chosen: by an appropriate pretreatment the GaAs(0 0 1) surface should be sufficiently depleted of As in order to avoid the formation of iron arsenides. Also, the growth of the Fe layers should be done at sufficiently low temperature to avoid intermixing. The intention of the present study was to check if epitaxial growth can be obtained under these conditions without the formation of 'dead layers'. For this purpose we have grown ultrathin Fe films on GaAs(0 0 1) surfaces by molecular beam epitaxy (MBE) and magnetron sputtering and investigated their structure, magnetic moments and in-plane magnetic anisotropy.

2. Experiment

Substrate preparation and film growth were done in an MBE chamber with a base pressure below $10^{-10}$ mbar. Commercial GaAs(0 0 1) wafers were cleaned in boiling isopropyl alcohol and introduced into UHV without further chemical treatment. The subsequent substrate surface treatment consisted of annealing in UHV at 500–550°C followed by Ar ion sputtering ($E_{\text{ion}} = 500$ eV) at the same substrate temperature and a final UHV annealing. Chemical composition and surface structure were repeatedly checked by Auger electron spectroscopy (AES), low-energy electron diffraction (LEED), reflective high-energy electron diffraction (RHEED) and in situ scanning tunneling microscopy (STM). In particular, RHEED turned out to be the most useful technique because it allows to continuously monitor the surface structure during the whole preparation and deposition process.

After the specific pretreatment of the substrates Fe was deposited at a rate of 1–2 ML/min (monolayers per minute). Changes of the surface structure and morphology were monitored by RHEED and STM. Finally, a covering layer of 20–50 ML Au was deposited for protection against corrosion. During film growth the pressure in the chamber is typically around $2–3 \times 10^{-10}$ mbar.

In a second experiment Fe films were grown on GaAs(0 0 1) in a UHV-based magnetron sputtering system with a growth rate of 30 ML/min after the same in situ annealing and ion etching procedure as for the MBE films. For these samples film structure was characterized ex situ by X-ray diffraction and STM.

3. Growth and structure

GaAs surfaces did not show any electron diffraction pattern prior to annealing and etching; AES revealed a large amount of oxygen and carbon. Oxygen was removed by annealing but carbon only disappeared after ion etching. Fig. 1 shows the RHEED pattern of a clean GaAs surface after annealing and sputtering. The diffraction spots form a Laue circle typical for an atomically flat surface. The symmetry of the pattern indicates the presence of the [1 1 0] direction.
of a $p(4 \times 6)$ surface reconstruction which means a considerable As depletion compared to the As terminated surface. In Fig. 2 the stripe pattern of a partially reconstructed GaAs surface is seen in the STM image. It should be noted that the orientation of the stripes is uniform over the whole sample on all the terraces which is due to the inherent anisotropy of the GaAs(0 0 1) surface due to the dangling bonds.

When Fe is deposited by MBE at a substrate temperature of about 300 K RHEED patterns indicate the formation of three-dimensional nuclei followed by a gradual smoothening of the Fe film. Coalescence occurs at 3–4 ML of Fe. During deposition of the Au protective layer the smoothening proceeds much more pronounced. The Au surface exhibits a $p(2 \times 2)$ reconstruction due to the presence of segregated As [6]. After slight sputter etching and annealing the 5-fold reconstruction of the clean Au(0 0 1) surface becomes visible.

Both RHEED and LEED diffraction patterns indicate good epitaxial growth of Fe and Au despite the low growth temperature. The Fe and Au (0 0 1) planes are parallel to the GaAs(0 0 1) surface. Within the plane we find the relative orientations $\text{Au}[1 0 0]||\text{Fe}[1 1 0]||\text{GaAs}[1 1 0]$ as expected.

This corresponds to a lattice misfit of 1.5% for Fe on GaAs(0 0 1) and of 0.5% for Au on Fe(0 0 1).

The orientation of sputtered films was investigated by X-ray diffraction. Fig. 3 shows a so-called $\phi$-scan of a 50 nm Fe film obtained by orienting the sample to a (1 0 3) diffraction spot of Fe or a (2 2 4) diffraction spot of GaAs and subsequent rotation around the surface normal. The spot widths of 3.8$^\circ$ for GaAs and 4.9$^\circ$ for the Fe layer indicate excellent epitaxial growth.

4. Magnetic moments

The total magnetic moments of the films were measured with a SQUID magnetometer (superconducting quantum interference device) in a wide range of temperatures ($10 \text{ K} \leq T \leq 400 \text{ K}$) and applied magnetic fields ($H \leq 5 \text{ T}$). For each temperature the magnetic moment as a function of applied field within the film plane is extrapolated back to $H = 0$ from the saturated linear part of the magnetization curve at high fields. The spontaneous moment obtained in this way is plotted versus temperature. It can be fitted well with a $T^{3/2}$ law even for the thinnest films as long as they are continuous [7]. From this fit according to

$$m_s(T) = m_0(1 - BT^{3/2}),$$

Fig. 2. Scanning tunneling micrograph of a GaAs(0 0 1) surface which shows the RHEED pattern of Fig. 1. The stripe pattern along [1 1 0] shows the 4-fold periodicity along [1 1 0]. The image displays four different terrace levels separated by monoatomic steps.

Fig. 3. X-ray diffraction $\phi$-scan of a 50 nm sputtered Fe film on GaAs(0 0 1) by selecting the Fe(1 0 3) and GaAs(2 2 4) spots and rotating the sample around the film normal (Cu-K$_\alpha$ radiation). From the spot width of 3.8$^\circ$ for GaAs and 4.9$^\circ$ for Fe excellent epitaxial growth is concluded.
we obtain the spin wave parameter, $B$, which is not discussed in this paper, and the ground state magnetic moment, $m_0 = m_0(T = 0)$.

In order to determine the average magnetic moment per Fe atom, we need the total number of Fe atoms in the film in addition to the sample moment, $m_0$. After a careful analysis of various methods, we chose X-ray fluorescence spectroscopy to determine mass-equivalent film thicknesses in the monolayer range for several reasons: it is non-destructive, repeatable, highly reproducible and has sub-monolayer sensitivity. By using secondary target excitation and filtering (e.g. Zn secondary target, Ni filter) the background intensity in the spectrum is drastically reduced so that the thickness of a 1 ML Fe film can be measured with an error less than 2% within a reasonable data accumulation time ($\approx 1$ h) as seen from Fig. 4. Calibration is achieved by using thicker films with their mass determined by a high-precision microbalance. In addition, the results are corrected for absorption and enhancement effects which account for different covering layers and substrates. Details of the method are discussed elsewhere [8].

For a 7 ML Fe film MBE grown on a GaAs(0 0 1) surface with a $(4 \times 6)$ reconstruction we obtain an average ground state magnetic moment per Fe atom of 2.3 $\mu_B$. In order to estimate the Fe moments in the immediate vicinity of the Fe/GaAs interface we make the following assumptions based both on results of ab initio band calculations [9] and experimental data [10]:

1. the Fe moments of the 5 inner layers of the 7 ML Fe film have approximately the bulk ground state moment of 2.2 $\mu_B$.
2. the Fe moments at the interface layer with the Au(0 0 1) covering layer are the same as the interface moments measured for epitaxial Fe films grown on Au(0 0 1) and covered by Au(0 0 1) [11] as shown in Fig. 5. The value of 2.9 $\mu_B$ per interface Fe atom determined from these data agrees well with theoretical predictions [12].

With these assumptions the Fe moments at the GaAs interface result to be 2.1 $\mu_B$ per atom which practically equals the bulk BCC-Fe moment. The room temperature moments are approximately 10% smaller which is explained by enhanced spin wave excitations in ultrathin films.

In Fig. 6 the total sample moment of Au(0 0 1)/Fe(0 0 1)/GaAs(0 0 1) films is plotted versus Fe film thickness for $T = 0$ and $T = 295$ K. The absence of any ‘dead layers’ at both temperatures is evident. On the contrary, a slight enhancement of the interface moment can be concluded from the positive extrapolated ground state moment for $t = 0$ (or negative ‘dead layer’ which would mean a ‘live layer’). At room temperature the ‘dead layer’ is

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Fig. 4. X-ray fluorescence spectrum of a 3 ML Fe(0 0 1) film on GaAs(0 0 1) measured with a Si(Li) detector, Zn secondary target excitation and Ni filter (raw data without any background subtraction). The amount of Fe is determined from the area under the Fe-K$_\alpha$ line. A sensitivity of a small fraction of one monolayer is achieved.

Fig. 5. Average magnetic moment per Fe atom of epitaxial Fe(0 0 1) films between Au(0 0 1) as a function of film thickness, $N_{Fe}$, in monolayers (ML) normalized to the bulk moment of 2.2 $\mu_B$. The solid line represents results from ab initio band calculations [9, 12]. The data are obtained by first extrapolating magnetization curves back to zero field and then by extrapolation to zero temperature [11].
Fig. 6. Spontaneous magnetic moments of MBE grown Fe(0 0 1) films as a function of Fe layer thickness. The absence of 'magnetically dead layers' is clearly seen for $T = 0$ and $T = 290$ K. The slope of the data at $T = 0$ corresponds to the Fe bulk magnetization.

equivalent to 0.06 ML which is explained by spin wave excitations as mentioned above.

This means that by room temperature growth on As depleted GaAs(0 0 1) surfaces we have been able to combine good epitaxial growth with full Fe moments at the interface. This is in strong contrast to previous publications where from 5 to 40 'dead layers' have been observed at 295 K for a growth temperature of 323 and 523 K, respectively [3], or a strong decrease of the average magnetization for Fe films thinner than 10 nm [4]. This result indeed is encouraging with respect to the possibility of injecting spin polarized electrons from a ferromagnetic metal into a semiconductor. Further experiments including film deposition at different temperatures and post deposition annealing are currently carried out.

5. In-plane magnetic anisotropy

An alternating gradient magnetometer (AGM) was used to determine the in-plane magnetic anisotropy of Fe films grown on GaAs(0 0 1). For thicker films we observe the expected 4-fold anisotropy of BCC-Fe as seen from a polar plot of the magnetizing energy, $W_{mag} = \int_0^\phi \mathcal{H} \, d\mathcal{M}$, for a 350 ML thick Fe(0 0 1) film grown by sputtering (Fig. 7); however, a small uniaxial contribution is already visible here. This becomes dominant if we reduce the thickness to 7 and 5 ML as seen in the hysteresis loops of Fig. 8. Now the easy axes of the bulk cubic anisotropy, (1 0 0), have become intermediately hard, [1 1 0] is the resulting easy and [1 1 0] the net hard axis. It should be noted that the absolute saturation values of these hysteresis loops as a function of film thickness confirm the absence of any dead layers at room temperature as was already stated above.

The anisotropy field of this uniaxial term in the 7 ML film, $H_K = 880$ Oe, is much stronger than that of the bulk cubic term. The 4-fold term has practically disappeared as shown in the polar plot of the magnetizing energy for 7 ML Fe in Fig. 9. This is reminiscent of the sign reversal of the 4-fold in-plane anisotropy constant, $K_1$, observed in Fe films grown on Au(0 0 1) which vanishes for the same thickness of 7 ML [13]. It will be checked by systematic measurements whether the reduced value of $H_K$ for the 5 ML film compared to 7 ML and the slight S shape of the hard axis loop indicated in Fig. 8 indeed means a sign reversal of $K_1$. It would be surprising if $K_1$ vanished at the same thickness for Au/Fe/GaAs(0 0 1) as for Au/Fe/Au(0 0 1) because this behaviour results from the competition between the bulk and the
interface fourfold anisotropies with the latter expected to be different for different interfaces.

A uniaxial magnetic anisotropy has already been observed before in thicker Fe films on GaAs(0 0 1) [2, 3]. There have been speculations on the origin of this anisotropy, e.g. whether it is connected with the presence of a Fe$_2$Ga$_{2-x}$As$_x$ at the interface [3]. Our results, however, support the explanation of the uniaxial term by the intrinsic anisotropy of the dangling bonds at the GaAs(0 0 1) surface which was suggested earlier [2, 4, 5]. This anisotropy could be useful for designing future magneto-electronic memory or switching elements.

6. Conclusion

We have grown epitaxial Fe films on GaAs(0 0 1). By Ar ion sputtering and UHV annealing clean and atomically flat GaAs(0 0 1) surfaces have been prepared. Good epitaxial growth was achieved both by MBE and magnetron sputtering with the substrate held at room temperature. This is confirmed by the in-plane magnetic anisotropy of the Fe films. Magnetic measurements showed that films MBE grown at room temperature on Ga-rich surfaces do not have any 'magnetically dead layers' at $T = 0$ nor at $T = 300$ K. This result is in contrast to previous studies where a drastic reduction of the average magnetization has been found in Fe/GaAs(0 0 1) films thinner than 10 nm [4] which corresponds to a 'dead layer' between 5 and 40 ML at the interfaces [3]. It also may open the way to realize electronic devices based on spin-dependent transport by injection of spin-polarized electrons into a semiconductor. To approach this goal the next step must be to determine the spin polarization of injected electrons and its dependence on the band structure of different metal electrodes, the Schottky barrier height and other relevant parameters.
References