Direct Observation of a Bulklike Spin Moment at the Fe/GaAs(100)-4 × 6 Interface

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We have used x-ray magnetic circular dichroism, which offers a unique capability to give element specific information at submonolayer sensitivity, to determine the spin and orbital magnetic moments at the Fe/GaAs(100) interface. The wedge samples, grown by molecular beam epitaxy at room temperature, consisted of 0.25–1 monolayer (ML) Fe on GaAs(100)-4 × 6 capped with 9 ML Co and have shown Fe spin moments of (1.84–1.96)μ_B and a large orbital enhancement. Our results demonstrate unambiguously that the Fe/GaAs(100)-4 × 6 interface is ferromagnetic with a bulklike spin moment, which is highly promising for spintronics applications.

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Interest in the Fe/GaAs interface has been largely spawned by continuing research aimed at finding suitable ferromagnetic metal–III-V semiconductor combinations for use in next-generation spintronic devices. At present, semiconductor device technology is able to make use only of the presence or the absence of charge, whereas future spintronic devices will utilize an extra degree of freedom, that of electron spin, which is most clearly seen in magnetic materials. In order that both technologies can coexist and, indeed, interface with each other, it is necessary to promote spin transport between semiconductor and magnetic surfaces [1–5].

An Fe/GaAs structure should, in principle, naturally lend itself to this kind of device as the lattice mismatch between Fe and GaAs is very small compared to other transition metal–III-V semiconductor combinations and because GaAs is the most popular III-V semiconductor in use today. High quality epitaxial growth of Fe on GaAs has been achieved by several groups [6–13], since the lattice constant of 2.87 Å for Fe is almost exactly equal to half that of GaAs, 5.65 Å [14]. However, until recently it was thought that Fe/GaAs structures would not be suitable for this kind of application for two key reasons: (i) the high conductivity mismatch between the two systems [15]; and (ii) the supposed presence of magnetically dead or half-magnetic layers at the interface between 0.7 nm [7,8] and 3.3 nm [6] in thickness [6–8,16]. The issue of the conductivity mismatch could have proved fatal as it would have caused poor spin transport between the two systems. This issue has recently been put to rest by the discovery of Schottky barrier effects at the interface [17–20], which would lead to an enhancement of the spin polarized transport across the interface.

The question over the presence, or otherwise, of magnetically dead layers has, for many years, been a hot topic of debate. Originally, dead layers were thought to exist due to a combination of factors: the interdiffusion of As with Fe and the growth morphology. Such seemingly insurmountable barriers have led some groups to abandon the Fe/GaAs system in favor of other systems such as Fe/ZnSe(001) [3]. However, as long ago as 1998 it was experimentally shown that the evolution of the ferromagnetic phase is primarily determined by the growth morphology in successive thicknesses [11] as opposed to being dominated purely by reacted phases at the interface [3]. Using *in situ* magneto-optical Kerr effect measurements, Xu *et al.* [11] were able to show a transition from a superparamagnetic to ferromagnetic phase at around 4.8 monolayers (ML) Fe where the structure begins to change from large islands towards formation of a continuous film.

Further work has found no evidence of dead layers for samples of 7 ML Fe/GaAs at room temperature [10,13], and previous investigations of ultrathin Fe (8 ML) and (33 ML)/GaAs(100) films by x-ray magnetic circular dichroism (XMCD) [21] demonstrated a stable spin moment approaching the interface and a giant enhancement of the orbital moment in the same region. For thinner samples As bonding issues complicate study and though ferromagnetic long-range order and uniaxial magnetic anisotropy exist at low temperatures for a critical thickness of 2.5 ML Fe/GaAs [12,22], the impossibility of such a system having a Curie temperature in excess of room temperature has previously been a considerable barrier in attaining information useful to the development of future spintronic devices.

To measure magnetism at interface levels provides a special challenge in so far as in the island growth regime, below around 5 ML, it becomes very difficult to control the behavior of the evaporated Fe with respect to the As

atoms of the substrate. Recent theoretical results [23,24] confirm that if the Fe atoms become covered by a floating layer of As atoms just 1 ML thick or if each Fe atom is bonded to two or more As atoms the Fe magnetic moment would be quenched. Hence, to retain a bulklike magnetic moment the Fe atom must be close to at least one other of its own kind. These problems would prove to be an insurmountable barrier were one to simply construct a submonolayer of Fe on even Ga-rich GaAs as there would simply not be enough Fe atoms present to counteract the effects of the As present. This would also seem to support recent results [12], suggesting that the onset of magnetism may not be directly related to the GaAs reconstruction. To overcome As bonding problems we have grown a 9 ML thick epitaxial layer of Co immediately on top of submonolayer Fe films to disperse the floating As layer clear of the Fe atoms. The presence of the Co also serves to provide the Fe with a source of exchange interaction, allowing very low thicknesses to become magnetic at room temperature by effectively simulating the behavior at the interface of a ferromagnetic Fe film. Using the element specific technique of XMCD [25,26], which is sensitive to depths up to 5 nm, we were able to study the magnetic characteristics of the Fe buried layer.

The wedge-shaped Fe films were grown by molecular beam epitaxy on a commercially grown substrate of GaAs(100) topped with a smooth epilayer and capped with As. Prior to growth, the substrate was annealed initially at 340 °C to desorb the As capping layer from the surface before further annealing at 550 °C for 1 h to promote a clean and ordered Ga-rich surface. Once the system had cooled to room temperature, the Fe was deposited onto the surface at a rate of approximately 1 ML per minute. A shutter was drawn across the sample to create the distinct thicknesses of the wedge. The time intervals between shutter movements were defined by the deposition rate indicated by a quartz microbalance, calibrated using reflection high-energy electron diffraction oscillations. After the desired thicknesses were achieved, the samples were capped off with 9 ML Co to limit the influence of As bonding followed by 7 ML Cr to enable easy transfer to the synchrotron facility. The thicker films examined, 4 ML Fe and above, were grown in the same manner, but with a Cr capping layer only [11].

XMCD measurements were performed on station 1.1 of the Synchrotron Radiation Source at Daresbury Laboratory (U.K.). The station was equipped with a superconducting magnet in order to apply a field of 3 T parallel or antiparallel to the incoming x-ray beam. Measurements were made at room temperature and with the 75% circularly polarized x rays incident perpendicular to the sample surface (cf. Fig. 1). The resulting current output from the sample was measured in total electron yield mode as a function of the photon energy. The dichroism was obtained as the difference spectrum, $I^+ - I^-$, achieved by reversing the direction of the applied magnetic field at fixed polarization.

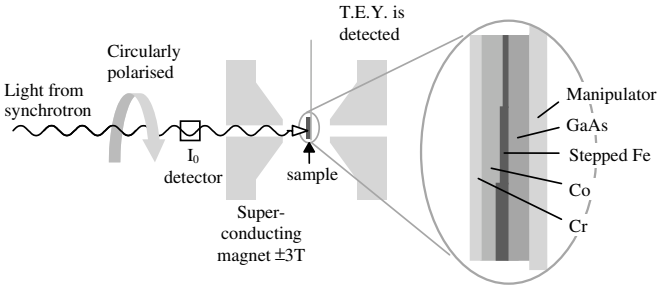

FIG. 1. A schematic diagram of the XMCD setup and sample structure (not to scale).

The traces shown in Fig. 2 represent the normalized x-ray absorption spectra (XAS) spectra taken at opposite magnetization directions and the resulting XMCD spectra for the three island growth mode thicknesses measured 1, 0.5, and 0.25 ML, designed to describe the magnetic moments towards the interface. From this figure we can see the samples are free from oxidization and display large differences between the positive and negative applied fields even for extremely low Fe coverage. The pre-edge structure shown in the 0.25 ML XAS is that of the Cr L_1 edge that is visible only against the background due to the extremely low level of Fe present and plays no part in the corresponding XMCD spectrum.

As a complement, Fig. 3 shows three pairs of normalized XAS spectra and corresponding XMCD traces for three thicknesses, without Co over layers, chosen to illustrate the evolution of the continuous film morphology, from 4 ML at the end of the superparamagnetic phase through to 8 and 18 ML in the ferromagnetic region.

The data were analyzed by applying the XMCD sum rules to the $2p$ to $3d$ spectra [27,28], using the procedure described by Chen *et al.* [29]. In this procedure, subsequent to the removal of a fitted, stepped background, integrals were taken of the XMCD signal $I^- - I^+$ and the sum signal $I^- + I^+$. The orbital and spin moments are obtained as

$$m_l = \frac{-4qn_h}{3rP}, \tag{1}$$

$$m_s = \frac{n_h(4q - 6p)}{rP}, \tag{2}$$

respectively, where p is the integral of the XMCD intensity over the L_2 edge, q is the integral of the XMCD intensity over the L_2 and L_3 edges, and r is the integral over the sum intensity of the L_2 and L_3 edges. The latter is required for the normalization and relates to n_h, the number of holes, which is taken to be 3.39 for Fe [28]. The degree of polarization, P, is equal to 0.75 for the experiment. Processing the data in this manner allows the results to be presented on a per atom basis.

Applying these techniques to the XMCD results of the submonolayer thicknesses in Fig. 2, we were able to identify the presence of a bulklike spin moment, $m_{s,\mathrm{bulk}} = 1.98\mu_B$ [29], finding values of $m_s = (1.84 \pm 0.11)\mu_B$,

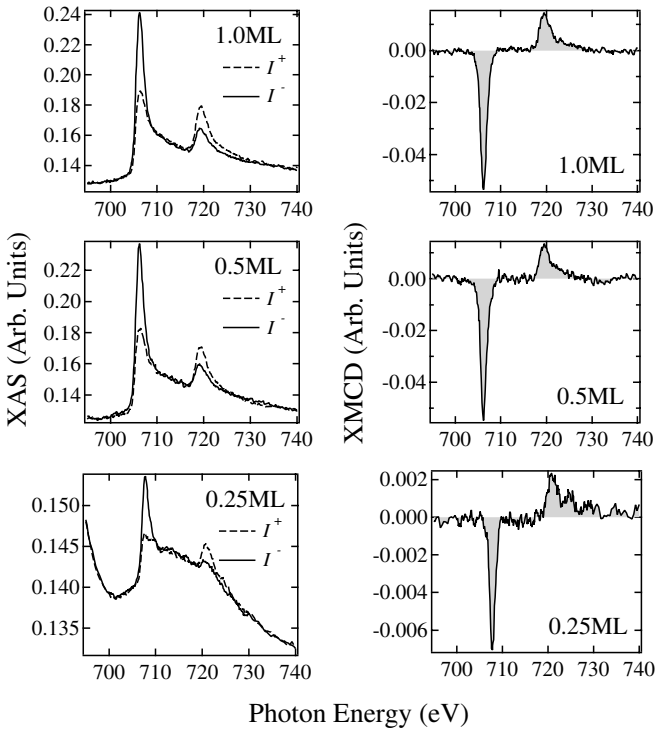

FIG. 2. Normalized XAS spectra, $I^{\pm}$, and corresponding XMCD spectra for Fe thicknesses of 1, 0.5, and 0.25 ML capped with 9 ML Co followed by 7 ML Cr.

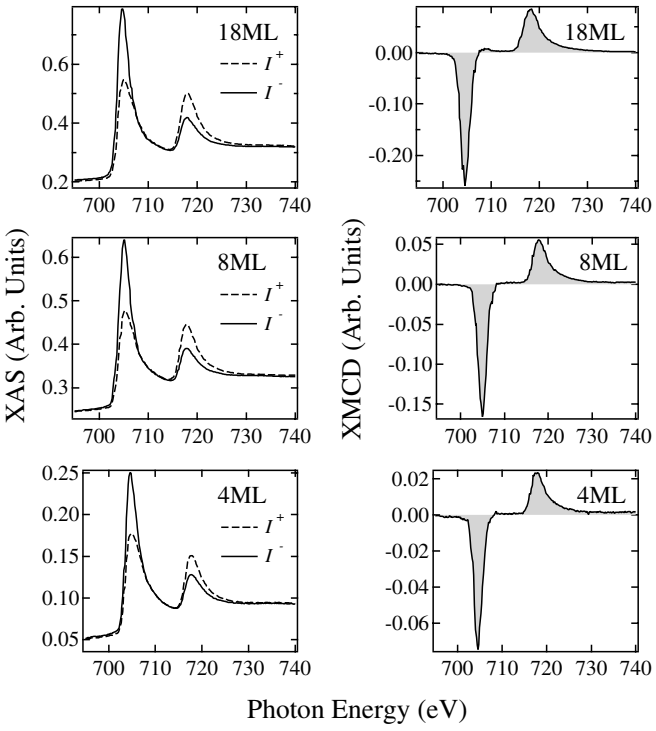

FIG. 3. Normalized XAS spectra, $I^{\pm}$, and corresponding XMCD spectra for Fe thicknesses of 18, 8, and 4 ML capped with 7 ML Cr only.

$(1.84 \pm 0.21)\mu_B$, and $(1.96 \pm 0.5)\mu_B$ for the thicknesses of 1, 0.5, and 0.25 ML, respectively. Analysis of the orbital moment revealed a giant enhancement of the orbital moment compared to the bulk value [29]. The values $m_l = (0.23 \pm 0.04)\mu_B$, $(0.25 \pm 0.05)\mu_B$, and $(0.23 \pm 0.1)\mu_B$, corresponding to the thicknesses of 1, 0.5, and 0.25 ML, respectively, were obtained, representing an enhancement of nearly 300% compared to the bulk value of $0.085\mu_B$ identified by Chen *et al.* [29] and comparing well with previous investigations at 8 ML Fe [21]. It is expected that this enhancement is due to the reduction of symmetry in comparison with the bulk structure and thus a change in the orbital degeneracy. Differences in comparison to the bulk structure are thought to originate from interdiffusion, steps, or terraces of the substrate, such as the Ga dimer row formed in the [011] direction of the GaAs(4×6) reconstruction [30]. This is thought to bring about localized $3d$ wave functions resulting in an enhancement of orbital moment [31].

From Fig. 3 the spin moments of the thicker films without Co over layers are $m_s = (2.21 \pm 0.04)\mu_B$, $(2.03 \pm 0.14)\mu_B$, and $(1.85 \pm 0.14)\mu_B$ for 4, 8 [21], and 18 ML Fe, respectively. These values are also close to that of the bulk, which further confirms the magnetic nature of the interface. We note that the spin moment for 4 ML is slightly enhanced. This enhancement is not yet fully understood but is thought to have its roots in the island structure present at this thickness. Our previous work has suggested Fe films on GaAs form superparamagnetic nanoclusters below around 5 ML, which as shown by Billas *et al.* [32] may demonstrate an enhanced magnetic moment due to the greatly reduced symmetry in these zero dimensional structures. Such a mechanism is consistent with results for single and submonolayer films, where the presence of the ferromagnetic Co overlayer would cause the moment enhancement associated with Fe nanoclusters to be circumvented. The values of the orbital moments, $m_l = (0.22 \pm 0.03)\mu_B$, $(0.26 \pm 0.03)\mu_B$, and $(0.15 \pm 0.02)\mu_B$ for 4, 8 [21], and 18 ML, respectively, show that there is still some reduced symmetry at 8 ML but that by 18 ML the system is becoming more bulklike with an orbital enhancement of 176%. This result is in keeping with a previous measurement of 33 ML [21], which gave an orbital moment of $m_l = (0.12 \pm 0.02)\mu_B$ and indicates that as the film thickness increases the symmetry rises towards that of the bulk state, and hence the wave functions become less localized. Reduction towards the bulk orbital moment does not appear to coincide with any change in magnetic phase, occurring well after the expected onset of the ferromagnetic phase that starts at ~4.8 ML.

Viewing the results of the submonolayer thicknesses in context with those obtained for thicker films, as shown in Fig. 4, clearly illustrates the presence of a bulklike spin moment and an enhanced orbital moment at the Fe/GaAs interface. These results show, in contradiction with previous reports [6–8,12,16,22], that islands of growth formed at the Fe/GaAs interface are individually

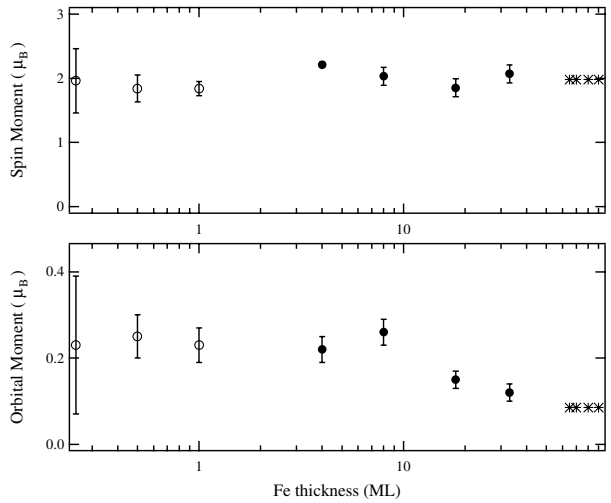

FIG. 4. The evolution of spin and orbital moment as a function of the Fe thickness. Open circles correspond to submonolayer films with Co overlayers, filled circles correspond to thicker Fe films, capped with Cr only, and the asterisks give the bulk values. The bulk values shown relate to results discussed in Ref. [29], while values corresponding to 8 and 33 ML Fe originate from Ref. [21].

ferromagnetic at room temperature when part of a larger ferromagnetic structure. Therefore, the results would appear to demonstrate that the risks of excessive As bonding at interface thicknesses, may be prevented where the interface is part of a larger ferromagnetic film. This is in agreement with the theoretical work of Erwin *et al.* and Merbt *et al.* [23,24], which suggests excessive As bonding at the interface to be a factor in the quenching of the magnetic moment. Thus, this result further establishes the Fe/GaAs system as a suitable candidate for the development of spintronic devices, which would necessarily have a Curie temperature in excess of room temperature.

In summary, ultrathin and submonolayer Fe films on GaAs(-4×6) surfaces have been studied with XMCD. This was aided by the inclusion of a Co stabilizing layer atop the monolayer and submonolayer Fe films, which facilitated the study of the spin and orbital moments at the Fe/GaAs interface. The measurements reveal a bulklike spin moment and a 300% enhancement in the orbital moment, showing that Fe at the GaAs interface is ferromagnetic. Similar results have also been found for the superparamagnetic thickness of 4 ML and ferromagnetic thickness 8 ML. The orbital moment enhancement may be characteristic of a loss of symmetry close to the interface, attributable to qualities of the substrate surface, resulting in localization of the Fe wave functions. The bulklike spin moment detected throughout the range of thicknesses studied demonstrated that the previously reported magnetic dead layer or half magnetic layer could be precluded and indicates Fe/GaAs is a suitable system for possible spin transport applications.

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