Giant enhancement of orbital moments and perpendicular anisotropy in epitaxial Fe/GaAs(100)

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The spin and orbital magnetic moments and the perpendicular magnetic anisotropy of 8 and 33 monolayer epitaxial bcc Fe films grown on GaAs(100)-4×6 have been measured using x-ray magnetic circular dichronism and polar magneto-optical Kerr effect. Both the films have approximately the same spin moments of about $2.0\mu_B$ close to that of the bulk value. The ultrathin film shows a giant orbital moment enhancement of about 300% with respect to the bulk value and a perpendicular interface anisotropy field $H_s^{\text{Fe-GaAs}}$ of the order of -5×10^4 Oe. This may be partially due to an increased degree of localization of electronic states at the Fe/GaAs interface associated with the atomic scale interface structure. © 2001 American Institute of Physics. [DOI: 10.1063/1.1359473]

INTRODUCTION

Interface magnetism in ferromagnetic metal (FM)/ semiconductor (SC) heterostructures continues to be an important topic for the study of fundamental magnetic properties of ultrathin films and for the development of next generation magnetoelectronic devices.^{1,2} There are two issues of current interest concerning Fe/GaAs,³⁻⁸ which is a model system for the study of FM/III-V SC heterostructures: first, the origin of the in-plane uniaxial magnetic anisotropy and second the possible existence of magnetic dead layers at the interface. We have shown in a previous study that it is the growth morphology rather than the FM SC intermixing which plays a dominant role in determining the evolution of the ferromagnetic phases with thickness and that there is no evidence of any significant dead layers forming at the interface in Fe/GaAs(100)-4 \times 6 grown at room temperature.⁷ In this article, we report a further study of this system using x-ray magnetic circular dichroism (XMCD) and polar magneto-optical Kerr effect (MOKE). The XMCD technique has been demonstrated experimentally and theoretically to be capable of determining both the spin and orbital magnetic moments with submonolayer sensitivity.9-17 We shall show that the Fe/GaAs interface has a full spin moment and that the ultrathin Fe films exhibit a giant orbital moment enhancement and a perpendicular magnetic anisotropy.

EXPERIMENT

Fe films were grown on GaAs substrates at ambient temperature (35 °C) in a molecular beam epitaxy chamber using an electron-beam evaporator. A buffer layer (~0.5 μ m) of homoepitaxial GaAs was grown on the commercial wafer to provide the smoothest possible GaAs surface. The As capped

^{a)}Corresponding address: Department of Electronics, The University of York, York YO10 5DD, United Kingdom; electronic mail: yx2@ohm.york.ac.uk substrate was transferred to a second growth chamber and then annealed to 550 °C for 1 h to obtain a clean and ordered Ga-rich 4×6 surface. Detailed growth and structure information can be found in Ref. 7. The samples were capped with 20 monolayers (ML) of Au for ex situ studies. The XMCD experiment was performed at beam line 1.1, CLRC Daresbury Laboratory, with 80% circularly polarized x rays. The Fe $L_{2,3}$ absorption spectra were collected at room temperature in total-electron-yield mode where the sample current was recorded as a function of photon energy. An external magnetic field of 3 T was applied perpendicular to the sample surface using a superconducting magnet. During the XMCD measurements the circular polarization of the photon beam was fixed and the magnetization direction was alternately aligned parallel/antiparallel to the propagation vector of the photon.

RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show the normalized x-ray absorption spectra (XAS) for the 8 and 33 ML films, respectively,



FIG. 1. (a) and (b) normalized XAS and (c) and (d) XMCD spectra of 8 and 33 ML films, respectively, with both the external magnetic field and the photon beam applied perpendicular to the film plane.

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FIG. 2. XMCD spectra of the 8 and 33 ML films scaled to 1 at the peak of the L_3 edge. The difference between the L_2 peaks is shown clearly in the inset.

with two opposite magnetizations. These two samples were chosen to represent an ultrathin film and a thin film, respectively, according to our detailed study of this system using in situ MOKE.⁷ The MXCD spectra shown in Figs. 1(c) and 1(d) are differences between the x-ray absorption spectra with the photon spin vector parallel (σ_{+}) and antiparallel (σ_{-}) to the sample magnetization corresponding to σ_{M} $=\sigma_{+}-\sigma_{-}$. The absolute value of the MXCD intensity at the L_3 edge, $|\sigma_M(L_3)|$, is proportional to the net magnetization. By normalizing $|\sigma_M(L_3)|$ to the total absorption cross section at L_3 , $\sigma_0(L_3) = 1/2 [\sigma_+(L_3) + \sigma_-(L_3)]$, the magnetization on a per atom basis can be obtained.¹³ We can see immediately from the figures that the ratios of the XMCD to the XAS are comparable in both films, indicating no reduction of the magnetic moment with decreasing thickness. To compare the XMCD spectra of two samples, both the spectra are scaled to 1 at the peak of the L_3 edge and are plotted in Fig. 2. This shows clearly that the dichroic area under the L_2 peak is smaller in the 8 ML film than that in the 33 ML film. Based on the XMCD sum rules^{10,12,17} that link the dichroic areas to the orbital and spin moments, this result suggests an orbital moment enhancement in the thin 8 ML film as compared with that of the 33 ML film.

The spin and orbital moments are determined by applying the XMCD sum rule¹⁰ with $n_h = 6.61$ (Refs. 10 and 15) and by subtracting a simple two-step background from the XAS spectra. It has been shown earlier that due to saturation effects in the total electron yield, the approximation $\sigma(\omega) \propto \omega Y(\omega)$ is not always valid for studies of magnetic films with photon incidence grater than 50°.⁹ We have chosen to work with 0° angle of light incidence with respect to the surface normal, so that the approximation $\sigma(\omega) \propto \omega Y(\omega)$ is valid to within 5%.⁹ As shown in Table I, the spin moments of the 8 and 33 ML films are $2.03 \pm 0.14 \mu_B$ and $2.07 \pm 0.12 \mu_B$, respectively, and the orbital moments are 0.26

TABLE I. Spin and orbital magnetic moments in units of μ_B /atom and saturation fields of two epitaxial Fe films on GaAs(100)-4×6.

	$m_{\rm spin}$	$m_{ m orb}$	$m_{ m orb}/m_{ m spin}$	$H_s(\times 10^3 \text{ Oe})$
8 ML	2.03 ± 0.14	0.26 ± 0.03	0.128	9±1
33 ML	2.07 ± 0.14	$0.12 {\pm} 0.02$	0.058	20 ± 2
bulk bcc Fe (Ref. 10)	1.98	0.085	0.043	21



FIG. 3. Magnetization loops of the 8 and 33 ML films with the field applied perpendicular to the film plane measured with polar MOKE.

 $\pm 0.03 \mu_B$ and $0.12 \pm 0.02 \mu_B$, respectively. The spin moments obtained are in agreement with our previous magnetic measurements.⁷ While the orbital moment of the 33 ML film is enhanced by about 40% compared with that of the bulk value, the 8 ML film shows a giant orbital moment enhancement of about 300%.

The giant enhancement of the orbital moment in the ultrathin films is expected to lead to an out of plane magnetic anisotropy. Figure 3 shows the magnetization loops of the 8 and 33 ML films with the field applied perpendicular to the film plane measured with polar MOKE. The magnetization of both films remains aligned in the plane, showing the dominant effect of the demagnetization field. However, the saturation field of the 8 ML film is significantly reduced as compared with that of the 33 ML film. As the spin moment of the films is independent of the thickness, the reduction of the saturation field clearly shows the existence of a perpendicular magnetic anisotropy. Neglecting a small volume anisotropy in Fe films,^{18,19} the saturation field H_s along the hard axis is determined by the following equation:

$$H_s = 4\pi M_s + (H_k^{\text{Fe-GaAs}} + H_k^{\text{Fe-Au}})/N, \qquad (1)$$

where $4\pi M_s$ is the demagnetization field, *N* is the number of the Fe atomic layers, and $H_k^{\text{Fe-GaAs}}$ and $H_k^{\text{Fe-Au}}$ are the interface anisotropy fields of the Fe–GaAs and Fe–Au interfaces, respectively. Taking $H_s = 9 \pm 1 \times 10^3$ Oe and $4\pi M_s = 2.1$ $\times 10^4$ Oe for the 8 ML film, the sum of $H_k^{\text{Fe-GaAs}}$ and $H_k^{\text{Fe-Au}}$ is found to be $-9.6 \pm 1 \times 10^4$ Oe. According to the previous studies of the epitaxial Au/Fe/Au systems by Heinrich *et al.*¹⁸ and Lugert *et al.*,¹⁹ the Fe–Au interface anisotropy field $H_k^{\text{Fe-Au}}$ is about -4×10^4 Oe.¹⁹ We would like to point out that the magnetization and the XMCD were both measured in large perpendicular fields at room temperature. Any reduction in M_s due to thermal effects is taken into account by measuring magnetic moments at room temperature. While a detailed thickness dependence study is needed to accurately determine the Fe–GaAs interface anisotropy field, the results from Fig. 3 show the existence of a perpendicular interface anisotropy in the order of -5×10^4 Oe at the Fe– GaAs interface.

In contrast with the bulk crystals where the orbital moment is nearly completely quenched, an enhancement of the orbital moment is expected at surfaces and interfaces due to a reduction of the symmetry, which can change the orbital

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degeneracy. First principle calculations on bcc Fe surfaces have predicted a 100% enhancement of orbital moment as compared to the bulk value,²⁰ which should be partially responsible for the observed orbital moment enhancement in the 8 ML film. However, the observed giant enhancement of about 300% suggests an additional mechanism. As pointed out by van der Laan et al.,¹⁷ the presence of surface roughness, interdiffusion, steps, or terraces will lead to more localized atomic-like 3d wave functions and an enhanced orbital moment. One of the distinct features of a reconstructed semiconductor surface is the formation of regular atomic scale structures, such as the Ga dimer row along the [011] direction in the GaAs(100)-4 \times 6 surface.²¹ We thus propose that this atomic scale structure leads to more localized wave functions for the Fe atoms close to the interface and a corresponding giant orbital moment enhancement and perpendicular surface anisotropy in the ultrathin film.

CONCLUSION

In summary, the spin and orbital magnetic moments and perpendicular anisotropy of the ultrathin epitaxial Fe films grown on GaAs(100)-4×6 have been studied using XMCD and polar MOKE. The spin moment of the films after the onset of the ferromagnetism was found to be $2.0\pm0.1\mu_B$ independent of the thickness of the films and close to the bulk value of $1.98\mu_B$.¹⁰ The orbital moment increases dramatically with decreasing thickness and is enhanced by about 300% with respect to that of the bulk bcc Fe in an 8 ML film. The polar MOKE measurements further suggest that the Fe–GaAs interface has a perpendicular anisotropy field of the order of -5×10^4 Oe. The giant orbital moment and the perpendicular anisotropy are attributed partially to an increased electron localization of the Fe atoms adjacent to the corrugated GaAs(100)-4×6 surface.

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