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Hybrid Fe_3O_4/GaAs(100) structure for spintronics

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Fe_3O_4/GaAs hybrid structures have been studied using reflection high-energy electron diffraction (RHEED), x-ray photoelectron spectroscopy (XPS), x-ray magnetic circular dichroism (XMCD), and low-temperature vibrating-sample magnetometry (VSM). The samples were prepared by oxidizing epitaxial Fe thin films in a partial pressure of 5×10^{-5} mbar of oxygen at 500 K for 180 s. Clear RHEED patterns were observed, suggesting the epitaxial growth of Fe oxides with a cubic structure. The XPS spectra show that the oxides were Fe_3O_4 rather than γ-Fe_2O_3, as there were no shake-up satellites between the two Fe 2*p* peaks. This was further confirmed by the XMCD measurements, which show ferromagnetic coupling between the Fe cations, with no evidence of intermixing at the interface. The VSM measurements show that the films have a magnetic uniaxial anisotropy and a "quick" saturation property, with the easy axes along the $[0\bar{1}1]$ direction. This detailed study offers further insight into the structure, interface, and magnetic properties of this hybrid Fe_3O_4/GaAs(100) structure as a promising system for spintronic application. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1857432]

I. INTRODUCTION

Magnetic oxides, such as Fe_3O_4 and CrO_2, have drawn intensive attention due to their half-metallicity and potential application in spintronics.[1] The iron oxide, Fe_3O_4, known as magnetite, has been prepared on Al_2O_3,[2] MgO,[2–5] and Pt (Ref. 6) substrates. But to integrate with the industry for spintronic application, the preparation of high-quality Fe_3O_4 on semiconductors, such as GaAs, is essential. At the same time, the Fe_3O_4 films grown by simultaneous oxidation upon evaporation, or dc reactive sputtering, have shown unexpected magnetic properties, such as superparamagnetism, remaining unsaturated even in the extra field as large as 70 kOe.[2] This slow saturation behavior has been attributed to the antiphase boundaries (APBs) appearing in samples with inverse spinel structure.[4] Does this unfavorable behavior persist for ultrathin Fe_3O_4, grown by postgrowth oxidation rather than by simultaneous oxidation? Another subject matter, to be clarified, is the interface and surface composition of the sample. When oxidized, how does the metallic Fe react with oxygen during the oxidation process at the GaAs surface? These problems are important for spin injection and the fundamental understanding of the oxide growth on semiconductors.

Our previous magneto-optical Kerr effect (MOKE) measurements of the Fe_3O_4/GaAs(100) hybrid structures have demonstrated the magnetic behavior, which is uniaxial in nature.[7] In this paper, we present a detailed study of the Fe_3O_4/GaAs(100) hybrid structure using reflection high-energy electron diffraction (RHEED), x-ray photoelectron spectroscopy (XPS), x-ray magnetic circular dichroism (XMCD), and low-temperature vibrating-sample magnetometry (VSM). The VSM measurement demonstrated that the slow saturation behavior is clearly absent. With XPS and XMCD, we provide further insight into the surface and interface characteristics of these structures.

II. EXPERIMENT

The magnetite samples were prepared by postgrowth oxidation of the pregrown epitaxial Fe on GaAs(100). The substrates were first chemically etched in a solution of H_2SO_4:H_2O_2:H_2O (4:1:1) for 30 s and rinsed in de-ionized water. After this the substrate was transferred to the growth/oxidation chamber, where a thermal annealing at 830 K for 30 min was performed, prior to growth. The Fe was held in a crucible and heated by an electron beam. The Fe growth rate was set to 2 Å/min, monitored by a quartz microbalance and the thicknesses were kept below 3 nm. The as-grown samples were then oxidized in an O_2 environment of 5×10^{-5} mbar at 500 K for 180 s. During the growth phase, RHEED measurements were made to monitor the crystal structure and morphology of the samples. The diffraction images were recorded with a charge-coupled device (CCD) camera.

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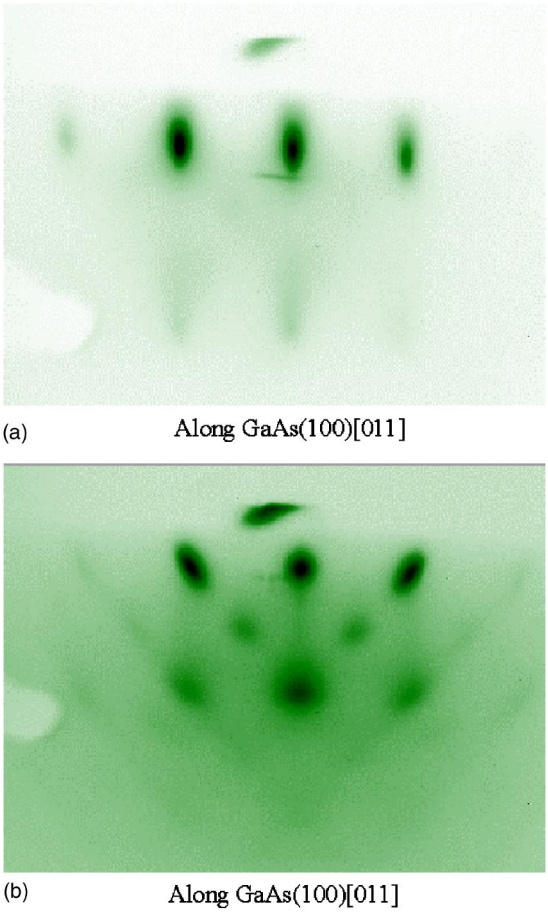

FIG. 1. RHEED images of the 3-nm Fe (a) and Fe_3O_4 (b) on GaAs(100) taken along the GaAs[011] direction. The high voltage of the RHEED gun is set to 10.0 kV.

When the oxide reaches a stable phase, showing no change in the RHEED pattern on further exposure to oxygen, the samples were taken out from the UHV chamber and exposed to air without any capping layer. VSM and MOKE measurements were carried out to detect the magnetic properties. Following this XPS measurements were performed with Mg $K\alpha$ radiation. XMCD measurements were carried out in station 1.1 of the Synchrotron Radiation Source at Daresbury Laboratory (U. K.). 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.

III. RESULTS AND DISCUSSION

The RHEED pattern of the 3-nm epitaxial Fe on GaAs(100) along the [011] direction is shown in Fig. 1(a). The RHEED pattern along the $[0\bar{1}1]$ direction (not shown here) is the same as Fig. 1(a), indicating a fourfold symmetry in the Fe crystal structure, consistent with the body-centered-cubic (bcc) structure. Shown in Fig. 1(b) is the RHEED pattern of the Fe oxide obtained along the GaAs(100)[011] direction. Simple calculation of the lattice parameter, according to the diffraction spots and comparison of the pattern with other reported RHEED images, is consistent with a Fe_3O_4 crystal structure.[5] However, the crystal structure of γ-Fe_2O_3 is very similar to the Fe_3O_4 and additional analysis was performed to examine the composition of the sample.

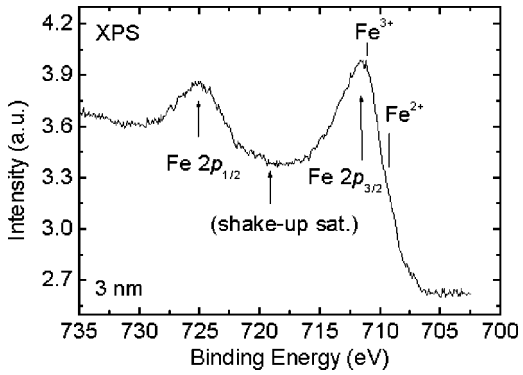

FIG. 2. The XPS core-level peaks of the 3-nm oxidized Fe. The Fe $2p_{1/2}$ and Fe $2p_{3/2}$ peaks are indicated by the arrows. Between these two $2p$ peaks, there is no satellite, which is the characteristics of Fe_2O_3, at the energy position indicated.

Verwey transition is believed to be a characteristic of the bulk Fe_3O_4. This is because of the freezing of the hopping electron between Fe cations upon cooling of the sample to around 120 K in the bulk Fe_3O_4 material. However, for ultrathin Fe_3O_4 films, the Verwey transition was found to be absent.[8,9] Therefore, rather than adopting this transition to characterize the oxides, we applied XPS, x-ray absorption spectroscopy (XAS), and XMCD to clarify the chemical states of the samples.

To study the surface composition of the structure, a 3-nm sample was measured with XPS and the Fe $2p$ core-level peaks are shown in Fig. 2. The main peak is visible at an energy of 711.0 eV, consistent with other reports.[10] The peaks are broader compared to the atomic Fe peaks. This can be explained by referring to the different chemical states of Fe cations presented in the sample. From the spectra another contributor can be identified, as marked in Fig. 2, with an energy of about 0.6 eV below that of the main peak. This peak is caused by the Fe^{2+} cations in the sample.[10] No shake-up satellite is detected in the spectrum. The shake-up satellite is the fingerprint of Fe_2O_3, and this satellite-free spectrum of the sample excludes the possible presence of γ-Fe_2O_3.

The XAS and XMCD measurements of a 2-nm sample are shown in Fig. 3 with an emphasis on the interface and surface of Fe_3O_4/GaAs(100) structure. In a previous work the Fe_3O_4/GaAs(100) structure was observed to display macroscopic-ferromagnetic behavior at around 2 nm (nominal thickness), measured by room-temperature MOKE.[7] In inverse spinel structure Fe_3O_4 crystal, one-third of all the Fe cations take up all the tetrahedral sites, and another one-third of the Fe cations fill half of the octahedral sites. These two kinds of Fe cations are antiferromagnetically coupled. The rest of the Fe cations inhibit the octahedral site. Here, clearly the contribution from the Fe^{3+}_{td} (Fe^{3+} cations at the tetrahedral sites) and Fe^{3+}_{oh} (Fe^{3+} cations at the octahedral sites) cancel each other out and the macroscopic ferromagnetism comes from the Fe^{2+}_{oh} cations. The shape of the XMCD spectra for

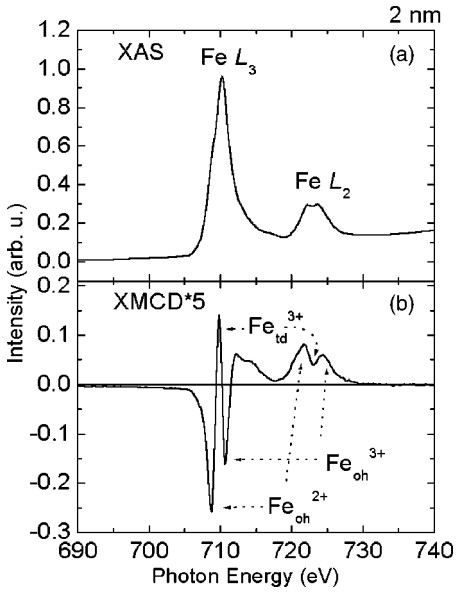

FIG. 3. The XAS (a) and XMCD (b) measurements of the 2-nm Fe_3O_4/GaAs(100) samples. The Fe_{oh}^{2+}, Fe_{td}^{3+}, and Fe_{oh}^{3+} peaks in the Fe L_3 and Fe L_2 edges are indicated.

the 2-nm sample is almost the same as the spectra obtained from other reports.[11] It was reported previously that at an elevated deposition temperature, Fe on GaAs(100) substrate reacts with As from the substrate and forms a nonmagnetic "dead" layer at the interface. But for the 2-nm sample, the XAS and XMCD spectra showed Fe_3O_4 characteristics and no evidence of the presence of any Fe–As compound.

Figure 4 depicts the low-temperature (150 K) VSM measurements of the 3-nm sample along the GaAs(100) [001] and $[0\bar{1}1]$ directions. Magnetic uniaxial anisotropy is clearly defined, with a ratio of M_r/M_s close to 1 and to 0 when the applied magnetic fields were along the $[0\bar{1}1]$ and [011] directions, respectively. The coercivity of the sample is around 250 Oe. Interestingly, the sample saturates quickly and the saturation field along the hard axis (the [011] direction of the GaAs) is around 900 Oe, which is greatly reduced compared to what is reported in the literature.[2] We attributed this reduction of saturation field to the decrease of APBs in the samples due to our preparation process. As the sample was prepared by postgrowth oxidation, the Fe_3O_4 first nucleates on the Fe surface and then grows. The Fe cations migrate from the Fe/oxide interface through the ionic oxide to the oxide/oxygen surface; the cations are added to the oxide which then thickens. Eventually, the whole ultrathin Fe film becomes oxidized.[12] In this scenario the Fe is quickly oxidized and grows epitaxially on the previously generated Fe_3O_4. While in the simultaneous oxidation, the oxidation might take place before the Fe atoms reach the substrate or the previously deposited oxides. In any event, the substrate or the deposited oxides are always oxygen rich due to the environment. So the APBs would be easy to develop in this simultaneous oxidation. However, in the postgrowth oxidation, the formation of the APBs is suppressed, as suggested by this study.

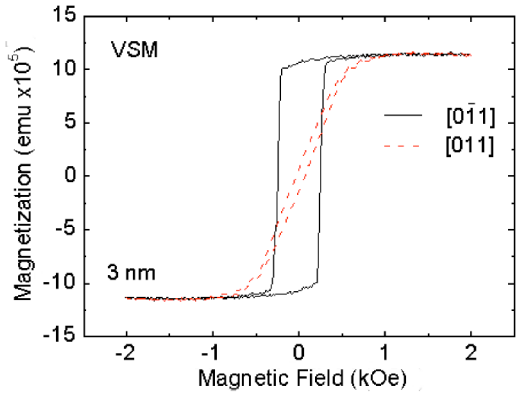

FIG. 4. Low-temperature VSM measurement of the 3-nm sample with magnetic field along the GaAs(100)[011] and $[0\bar{1}1]$ directions. The uniaxial magnetic anisotropy can be concluded from the different remnant ratio of the loops along these two directions.

IV. CONCLUSION

The magnetic properties of the surface and the interface of the Fe_3O_4/GaAs(100) hybrid structure have been investigated. Epitaxial Fe_3O_4 has been prepared on GaAs(100) by postgrowth oxidation. The surface of the sample is revealed to be Fe_3O_4 rather than Fe_2O_3. The Fe was found to react preferentially with the oxygen, by migration, rather than with As from the substrate. The XPS and XMCD measurements support this conclusion. The VSM loops indicate that the ultrathin Fe_3O_4 film unambiguously exhibits a quick saturation behavior with a saturated field of 900 Oe. The Fe_3O_4/GaAs(100) hybrid structures appear to be a promising candidate for spintronic devices.

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