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Annealing-Induced Fe Oxide Nanostructures on GaAs

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We report the evolution of Fe oxide nanostructures on GaAs(100) upon pre- and post-growth annealing conditions. GaAs nanoscale pyramids were formed on the GaAs surface due to wet etching and thermal annealing. An 8.0–nm epitaxial Fe film was grown, oxidized, and annealed using a gradient temperature method. During the process the nanostripes were formed, and the evolution has been demonstrated using transmission and reflection high energy electron diffraction, and scanning electron microscopy. These nanostripes exhibited uniaxial magnetic anisotropy. The formation of these nanostructures is attributed to surface anisotropy, which in addition could explain the observed uniaxial magnetic anisotropy.

***Index Terms*—Annealing, magnetic Fe oxide, magnetic uniaxial anisotropy, nanostructures.**

I. Introduction

FERROMAGNETIC nanostructures on semiconductors can lead to a new class of spin electronic devices based on the manipulation of the degree of freedom of spin [1]–[3]. One approach to the fabrication of these magnetic nanostructures is the "bottom-up" strategy:, i.e., to produce self-assembled nanoscale structures on specially treated substrates using molecular beam epitaxy (MBE), with the assistance of lattice mismatch induced strain and surface effects. This has been well demonstrated in heteroepitaxial semiconductor systems, for example, Ge [4] and InAs [5] nanostructures have been developed on Si and GaAs respectively due to their lattice mismatches.

Fe oxide is a promising candidate for spintronics, for example, Fe_3O_4 has high spin polarization at the Fermi energy level, with a Curie temperature above room temperature [2]. In a previous paper [6], we reported the very first synthesis of epitaxial Fe_3O_4 on GaAs(100) by post-growth oxidation. Furthermore, we have also reported that half-metallic Fe_3O_4 nanostripes can be generated on deformed GaAs(100) [7]. An anisotropic deformation of the chemically etched and annealed GaAs(100) substrate gives rise to an anisotropic stress/strain distribution in the deposited Fe_3O_4 layer, and nanostripes form during the oxidation process. The surface energy could also play a role in the formation of Fe_3O_4 nanostripes. In this paper, the annealing effects on the iron oxide nanostructure evolution from nanodots to nanostripes are presented. In addition, magneto-optical Kerr effect (MOKE) measurements reveal a uniaxial magnetic anisotropy (UMA) in these nanostructures, which could be explained in terms of the surface anisotropy of the GaAs substrate.

II. Experiment

The GaAs(100) substrate was first wet etched in a H_2SO_4:H_2O_2:H_2O (4:1:1) solution for 30 s.. After deionized water rinsing the substrate was fixed on a specially designed Ta sample holder, with a small window at its center. During annealing, the electron beam from the filament directly heats the back of the substrate through the small window, with the edge area contacted to the Ta sample holder. Due to thermal conductivity, the temperature of the edge area of the substrate is relatively low compared with the central part, and thus a temperature gradient is established over a distance of 5 mm with the temperature at the edge maintained at 870 K.

The oxide/GaAs(100) sample was grown by post-growth oxidation, as described in our previous work [6]. It should be noted that the substrate was fixed with its top surface facing the evaporation source, which was kept at a distance of 400 mm away from it. This configuration produces a film of uniform thickness. The Fe growth rate was set to 2 Å/min, as measured by a quartz microbalance. The epitaxial Fe was then oxidized by molecular O_2, with a partial pressure of 5×10^{-5} mbar and a temperature of 500 K at the substrate edge for 10 minutes. During the growth and oxidation phases, reflection high-energy electron diffraction (RHEED) patterns were observed in order to monitor the variation of the surface morphology.

Once the sample achieved a stable phase, as indicated by stable RHEED patterns upon increasing the dosage of oxygen or temperature, it was taken out of the MBE chamber. The surface morphology was then observed by a scanning electron microscopy (SEM). Finally MOKE measurements were performed to probe its magnetic properties.

III. Results

The RHEED patterns along the GaAs[0-11] direction of the annealed GaAs(100) prior to Fe growth and that of an 8.0 nm Fe sample are presented in Fig. 1(a) and (b), respectively. From Fig. 1(a), both the transmission and reflection patterns can be identified with a reconstruction in the reflection pattern. To some extent the patterns are streaky, suggesting a flat GaAs(100) surface, while the transmission dots indicating the presence of some nanostructures on the surface at the same time, through which the RHEED electrons were transmitted and diffracted. Fig. 1(b) is the Fe/GaAs RHEED pattern. Despite a 8.0-nm-thick Fe layer, the transmission pattern from the

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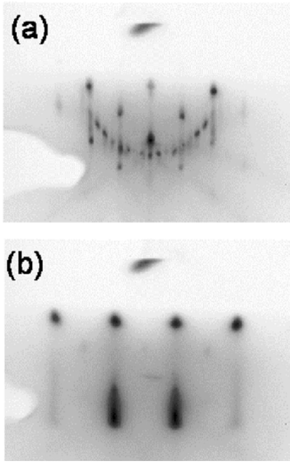

Fig. 1. (a) RHEED patterns of the GaAs(100) after annealing and of the 8.0–nm Fe on GaAs(100) along the [0-11] direction. The GaAs pattern is still visible despite the 8-nm Fe layer in (b). In (a), both transmission and reflection pattern are clearly demonstrated.

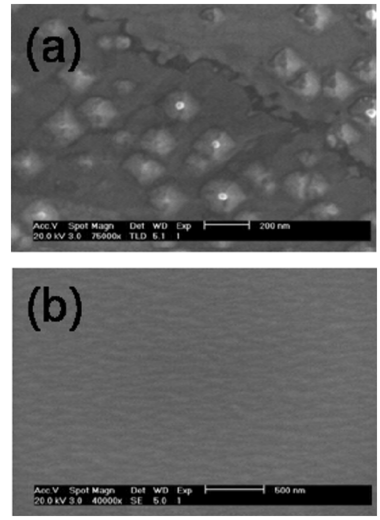

Fig. 2. SEM images of the annealed GaAs. (a) The centre area with pyramids. The edges of the pyramids are along the GaAs$\langle 001 \rangle$ directions. (b) Illustrates the uncovered GaAs(100) surface at the edge. The horizontal direction of the images are along the GaAs [011] axis.

GaAs(100) is still visible. This further supports the presence of the GaAs nanostructures, which form protrusions on the GaAs(100) surface and provide transmission pass-through for the incident RHEED electrons, producing the same pattern as bulk GaAs.

Prior to oxidation the Fe film was flat and no special structures had been found. RHEED patterns after oxidation reveal a structure of Fe_3O_4. However, we should note that due to the thickness of the Fe film (8 nm), the oxide is not uniform and a FeO layer might present at the interface, between Fe_3O_4 and the GaAs.

The formation of GaAs nanostructures is confirmed by SEM images, as shown in Fig. 2(a). Nanoscale pyramids form at the center of sample with the pyramid edges along the GaAs$\langle 001 \rangle$ directions. However, the edge area of the sample is relatively flat and no pyramid is observed, as shown in Fig. 2(b).

The formation of GaAs pyramids is intriguing. Circular Ga droplets introduced by over annealing have been found on the surface by other group [8]. This is believed to be due to the decomposition of the GaAs surface during over-annealing. However, in our work, rather than spherical droplets, pyramids with well defined square bases and the same edge directions, have been observed in the central region. We know nanoscale GaAs pyramids can not be produced in the wet etching process. Pyramid formation is thought to be due to the annealing process, which might have caused nonuniform stoichiometry in the surface. The spatial distribution of the pyramids at the centre of the substrate also suggests that the pyramid formation is related to the annealing process.

In the post-growth oxidation, another temperature gradient for oxidation has been established and the Fe oxide nanostructure evolution was realized, as shown in Fig. 3(a) and (b), representing the edge and the center of the sample, respectively. In Fig. 3(a), only the nanodots of the initial stage can be identified, as magnified in the inset. Whereas in Fig. 3(b), clear nanostripes are revealed, with the preferential elongation along the GaAs[011] direction. To some extent the nanostripes are irregular, with an average width of 10 nm.

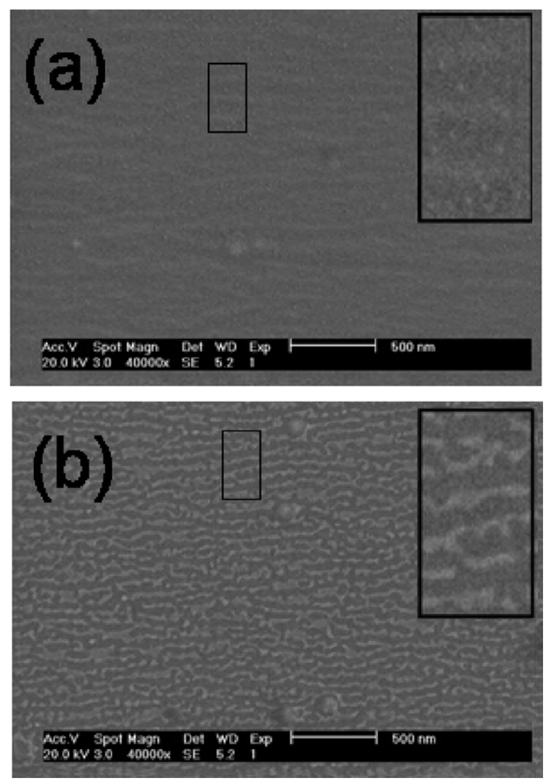

Fig. 3. SEM images. (a) The initial stage of nucleation near the sample edge. (b) Reveals the nanostripes formed near the center of the heating area which is at a higher temperature. The horizontal directions in the images are the [011] direction of the sample. The rectangle areas are magnified in the insets.

Typical MOKE measurements of the magnetic hysteresis loops from the nanostripes are shown in Fig. 4. In contrast to Fe film of the same thickness on GaAs, which exhibits a cubic magnetic anisotropy [9], the Fe oxide film displayed a uniaxial magnetic anisotropy, with the easy axis along the [0-11] direction and the anisotropy constant K_u around 1040 J/m^3, perpendicular to the preferentially elongated direction of the nanostripes.

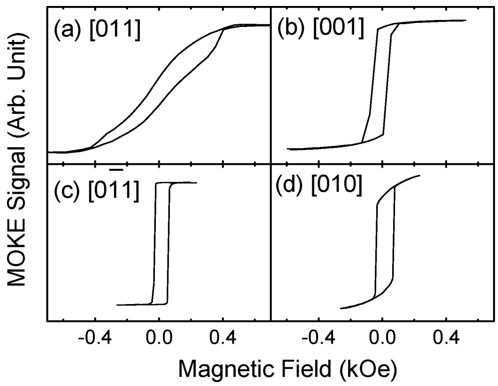

Fig. 4. Magneto-optical Kerr effect measurements along four directions of the sample. The direction of the applied magnetic filed are labeled.

This UMA easy axis is inconsistent with that of the shape anisotropy, which might be expected to be along the elongated direction of the nanostripes. So there are other contributions to the UMA, among which the stress/strain induced uniaxial anisotropy might dominate. At the same time, the contribution from chemical bonding can not be excluded. Further experiment would provide insights into the oxide/GaAs interface.

The GaAs surface anisotropy might induce this stress/strain-related UMA. Although the GaAs crystallizes in a cubic zinc-blende structure, the [011] and [0-11] directions are not identical and will introduce stress/strain anisotropy in the GaAs(100) surface and the layer above. The etching and annealing processes, prior to the sample preparation, enhanced the difference between the two directions. This difference introduces the UMA in the Fe oxide on GaAs(100), similar to the uniaxial magnetic anisotropy found in the Fe films on GaAs(100) [9].

IV. Conclusion

Annealing has been observed to be the driving force for the Fe oxide nanostructure evolution on GaAs(100). The nucleation and nanodots occurred at a relatively lower temperature near the sample edge, while nanostripes formed at a higher temperature around the center under direct e-beam heating. The preferential direction of the nanostripes is along the GaAs(100)[011] direction. The surface anisotropy of the GaAs might be the origin of the nanostripes as well as the uniaxial magnetic anisotropy, which is, surprisingly, perpendicular to the nanostripes.

Acknowledgment

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