Solid State Phenomena Vol. 213 (2014) pp 51-55
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doi:10.4028/www.scientific.net/SSP.213.51

The influence of seed layer on growth of magnetite films on the SiO_2/Si(001) surface

T.A. Pisarenko [a,1,2], V.V. Korobtsov [1,2], V.A. Vikulov [1], A.A. Dimitriev [1,2], V.V. Balashev [1,2]

[1] Institute of Automation and Control Processes, FEB RAS, 5 st. Radio, Vladivostok 690041, Russia

[2] Far Eastern Federal University, 8 Sukhanova St., Vladivostok 690091, Russia

[a]e-mail: tata_dvo@iacp.dvo.ru

Keywords: magnetite, seed layer, hybrid heterostructure, RHEED pattern, reactive deposition.

Abstract. Polycrystalline magnetite films with the thickness of 50 nm were grown on SiO_2/Si(001) surface by the reactive deposition of Fe in O_2 atmosphere using various preparation ways of the formation of iron oxide seed layer. The seed layers were formed by the deposition and oxidation of 3 nm Fe layer at different thermal conditions. It was found that polycrystalline magnetite films grown with the use of seed layer have [110] texture and are characterized by increase of the squareness of magnetic hysteresis loop. Structural analysis of magnetite films and predeposited seed layers was studied by RHEED.

Introduction

Hybrid heterostructures, integrating half-metallic magnetic oxides with semiconductor devices are important for the development of spintronics with high-efficient spin injection. Among the half metallic ferromagnets, magnetite (Fe_3O_4) has enormous attention due to its high spin polarization and high Cure temperature of 858 K [1].

Growth of Fe_3O_4 films on silicon substrates is very important because silicon is the most widely used material in modern semiconductor technology. However, at the initial stages of Fe_3O_4 film growth on Si(111) and Si(001) substrates the formation of iron silicide and amorphous oxide was found [2, 3]. That is detrimental for efficient spin injection from Fe_3O_4 into silicon.

Given the relatively large lattice mismatch between Fe_3O_4 and Si, a buffer layer is needed to grow Fe_3O_4 film on Si substrate. The crystalline structures of Fe_3O_4 films both directly deposited on Si [2, 4] and deposited on Si with buffer layers of Ta [5], Ti, Ta, SiO_2 [3], Cu [4], and Fe_2O_3 [6] is polycrystalline. In Ref. [3-6] reported that the presence of normal metallic buffer layer improves the crystallinity and magnetic properties of Fe_3O_4 films. It has been shown [7, 8] that a SiO_2 layer with a suitable thickness is a good buffer layer between Si and Fe_3O_4. However, in this case the formation of the magnetite film is carried out on the isotropic substrate, whereas fabricating of stoichiometric and high-crystallographic-quality Fe_3O_4 film is crucial for its promising application in the capacity of a highly polarized injector. In Ref. [9] shown that the 10 nm Fe_3O_4 seed layer, formed by oxidizing an ultrathin 5 nm Fe layer in an O_2 atmosphere at 260°C, effectively works as a template for the successive growth of 100 nm Fe_3O_4 film on GaAs by the reactive MBE deposition of Fe in O_2 atmosphere.

In this paper, we report on the similar effect of seed layer for the growth of Fe_3O_4 films on an SiO_2/Si(001) surface. It was found that the formation of the seed layer on the SiO_2/Si surface changes the film texture, and hence it changes the film magnetic properties.

Experiment

The experiments were performed in an ultra-high vacuum MBE-system "Katun" equipped with 20 keV reflection high-energy electron diffraction (RHEED) and ellipsometry instruments. The base pressure of the MBE system was 1×10^{-10} Torr. The n-type Si(001) wafers of 4.5 Ω×cm resistivity and 20×10×0.5 mm^3 in size were used as a substrates. Before the substrate loading into vacuum chamber an ultrathin SiO_2 layer was grown by boiling of Si substrate in nitric acid (HNO_3 68%) for 5 minutes. SiO_2 thickness was equal 1.5 nm according to ellipsometry. After loading in vacuum chamber the substrates were degassed at 500^{o}C for 1 hour. Iron was thermally evaporated from an alumina crucible with a rate of 0.8 nm/min. RHEED patterns were taken at a glancing angle of ~0.5-1.0^o.

Magnetite films with total thickness of 50 nm were grown on SiO_2/Si(001) surface by reactive deposition of Fe in O_2 atmosphere at substrate temperature of 300^{o}C both with iron oxide seed layer and without it. Generally, a 25 nm thick Fe was deposited to form 50 nm thick Fe_3O_4. The seed layers were prepared by two techniques: 1) 3 nm Fe layer was deposited onto SiO_2/Si(001) surface at room temperature (RT) and oxidized at temperature of 300^{o}C; 2) 3 nm Fe layer was deposited at temperature of 300^{o}C and oxidized at the same temperature. Oxidation of the seed layer as well as deposition of the remaining 22 nm Fe was carried out at the oxygen pressure of 2×10^{-6} Torr.

Results and discussions

In Fig. 1a RHEED pattern from the film grown without a seed layer is shown. This RHEED pattern is characteristic for the textured polycrystalline film.

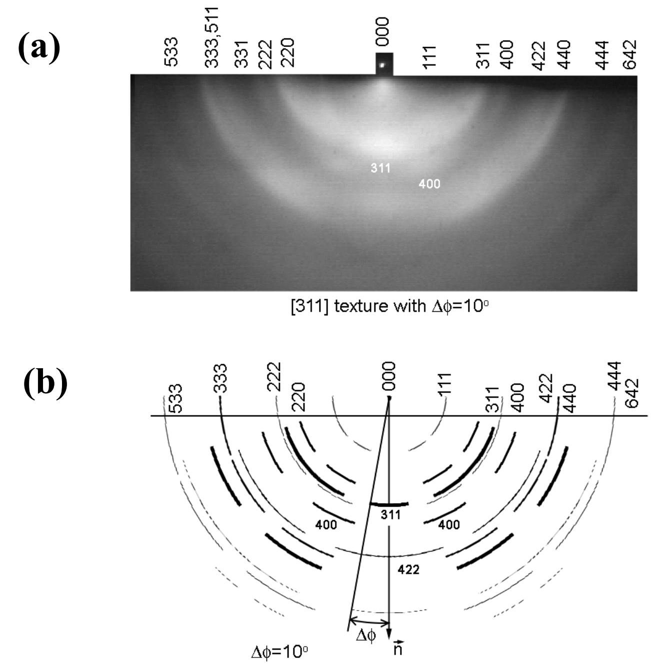

Fig. 1. *(a) RHEED pattern obtained upon depositing the 25 nm Fe layer at 300°C in an oxygen atmosphere and (b) the calculated transmission diffraction pattern for the polycrystalline Fe_3O_4 film with [311] texture.*

The positions of diffraction rings (Fig. 1a) agree with theoretical diffraction pattern calculated for a magnetite lattice with an inverse spinel structure. The texture in the magnetite film was analyzed by calculating the theoretical RHEED pattern using the kinematic approach [10, 11]. Analysis showed that the experimental RHEED pattern is best fitted by a theoretical diffraction pattern for a film with [311] texture (Fig. 1b). The axis of texture is parallel to normal **n** to the film surface. Offset Δφ of the [311] axis of the crystallite lattice from the normal to the surface is within ±10°.

Under the seed layer formation by the first technique the RHEED pattern exhibited diffuse rings after the deposition of 3 nm Fe layer on the SiO_2/Si(001) surface at RT. Then the sample was heated up to 300^{o}C. As result the sharpness of Fe rings increased and additional rings from iron silicide appeared as it was observed in our work [7] for the case of thick Fe layers. After the oxidation of the Fe layer for 10 minutes the diffraction rings from ε-FeSi remain and the diffraction rings from Fe transform to the diffraction rings from magnetite.

Sharpness of diffraction rings increases at the subsequent deposition of remaining 22 nm Fe layer in an oxygen atmosphere onto the seed layer results. In RHEED pattern (Fig. 2a) one can see that the film deposited onto the seed layer is also textured. However, the location of arcs indicates that the film has a texture other than the previous case. Comparison of the experimental (Fig.2a) with the theoretical RHEED pattern (Fig.2b) shows that the Fe_3O_4 film has [110] texture. The axis of texture is perpendicular to the film plane. Angular spread for the texture is equal to ±(10-15°).

(a)

(b)

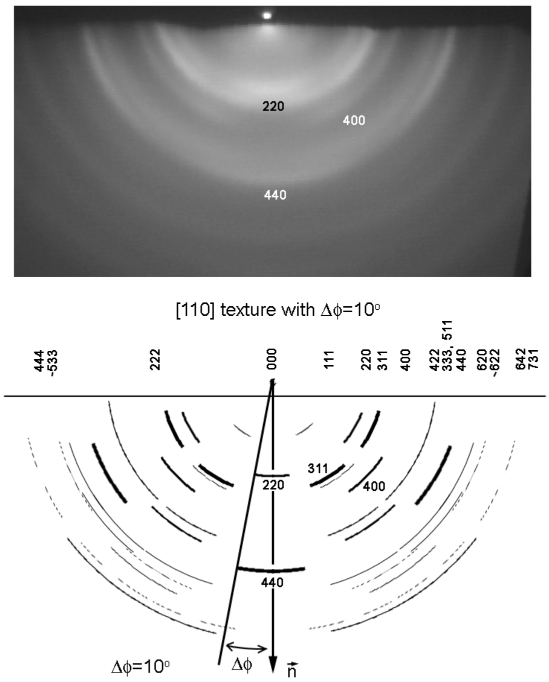

Fig. 2. *(a) RHEED pattern obtained upon depositing the 22 nm Fe layer at 300°C in an oxygen atmosphere on the seed layer and (b) the calculated transmission diffraction pattern for the polycrystalline Fe_3O_4 film with [110] texture .*

In the case of the seed layer formation by the second technique (the high temperature deposition of 3 nm iron layer) we observed only the diffraction rings from iron in the RHEED pattern. Oxidation of Fe layer at the same temperature resulted in appearance of Fe_3O_4 diffraction rings. The RHEED pattern of this seed layer characterized by more diffuse diffraction rings than the RHEED pattern of the seed layer formed by the first technique. This fact shows that the seed layer is a more fine-dispersed. However, the Fe_3O_4 film grown on this seed layer had the same texture as that in the case of the previous seed layer. This suggests that the procedures for the formation of seed layers used by us do not affect the texture. Nevertheless, the high temperature deposition of Fe for the seed layer formation is more preferable due to an absence of iron silicides.

We propose that the difference in the texture of the Fe_3O_4 films grown with and without the seed layer may be due to changes in the growth mechanism. In the absence of the seed layer on the SiO_2 surface the [311] texture formation occurs during growth. In addition, the initial stage of growth is nonstationary, which leads to the formation of an ultra-thin layer of non-stoichiometric composition on Fe_3O_4/SiO_2 interface. Whereas, the growth atop the seed layer excludes non-stationary stage of growth, and stoichiometric Fe_3O_4 grains of the seed layer determine [110] texture.

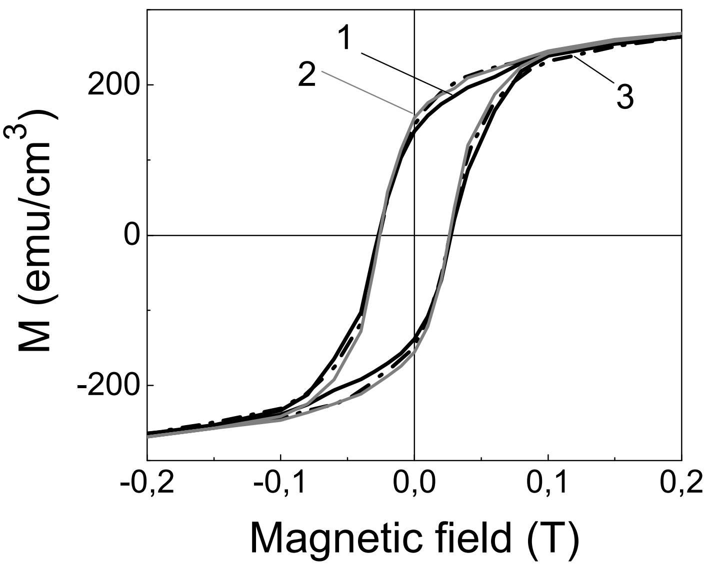

Fig. 3 *Magnetization hysteresis loops of the magnetite films: (1) without the seed layer, (2) with the seed layer grown by first technique, (3) with the seed layer grown by second technique.*

Magnetic measurements showed that the seed layer introduction influences not only on texture of the Fe_3O_4 films, but results in the changes of the magnetic properties of these films. Magnetization hysteresis loops for all samples are represented in Fig. 3. Analysis of these results has shown that the loop squareness for the sample without the seed layer is 10% less than that for the samples with the seed layer. The reason of the loop squareness increase under the seed layer introduction is attributed to the increase of magnetic coupling of grains as a structure quality becomes better.

Conclusions

The growth of magnetite films by Fe reactive deposition in an oxygen rich environment on SiO_2/Si(001) surface without and with the seed layer was investigated. The influence of the ultrathin iron oxide seed layer deposited on SiO_2/Si(001) surface on the growth mechanism was found. The Fe_3O_4 film formed by direct deposition on SiO_2/Si(001) has [311] texture, while the Fe_3O_4 films deposited atop the seed layer have [110] texture. Change of texture is explained by the exclusion of non-stationary growth stage. The seed layer formation by the deposition at 300°C and the oxidation at the same temperature is preferable because it eliminates the formation of iron silicide on Fe_3O_4/SiO_2 interface. By changing the oxidation method during the growth of the Fe_3O_4 initial layer, we fabricated stoichiometric and more crystalline quality Fe_3O_4 film. The formation of the half-metallic Fe_3O_4 films on to a seed layer allows to improve magnetic properties of the film and to control texture of the film making them more attractive for using in spintronic devices.

Acknowledgements

This work was supported by the Far East Branch of the Russian Academy of Sciences (project nos. 12-III-A-02-027 and 12-I-OFN-08), the Ministry of Education and Science of the Russian Federation ("Program in Support for Leading Scientific Schools," project no. NSh_774.2012.2), and the Russian Foundation for Basic Research (project No. 11-02-98523_r_vostok_a).

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