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The impact on the magnetic field growth of half-metallic Fe_3O_4 thin films

Xiao-Li Tang*, Huai-Wu Zhang, Hua Su, Zhi-Yong Zhong, Yu-lan Jing

Institute of Micro-Electronics and Solid-State Electronics, University of Electronic Science and Technology of China, Chengdu, Sichuan 610054, China

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Abstract

Half-metallic Fe_3O_4 films grown on a Si (100) substrate with a tantalum (Ta) buffer layer were prepared by DC magnetron reactive sputtering. Primary emphasis was placed on magnetic field growth of Fe_3O_4 thin film. The experiment's results showed that applying an external magnetic field to the samples during the growth was efficient to promote the polycrystalline Fe_3O_4 growth along certain directions. The magnetoresistance (MR) was also tested for comparison of the samples prepared with and without an external magnetic field, and showed that applying an external magnetic field can promote the MR values.

Keywords: Half-metal; Fe_3O_4; Spintronic; Magnetic film

1. Introduction

Presently, half-metallic films have attracted enormous attention due to their representing metallic properties for one electron spin population and an insulating property for the other. This property was first postulated by Rob de Groot et al. [1], and then several materials such as $La_{1-x}Sr_xMnO_3$ [2,3], NiMnSb [4,5], Sr_2FeMoO_6 [6], CrO_2 [7,8], and Fe_3O_4 [9–11] have been suggested as half-metallic ferromagnets. They are suitable candidates for application in spintronic devices due to their 100% spin polarization. Among them, more attention has been paid to Fe_3O_4, due to its high Curie temperature T_c (858 K) [12], as compared to other half-metallic films (such as LSMO: ~360 K, LFMO: ~430 K, CrO_2: ~395 K), and high conductivity, which relies on rapid electron hopping between Fe^{2+} and Fe^{3+} ions in the octahedral B sites of the inverse-spinel lattice under the room temperature.

Various techniques have been used to fabricate single-crystalline Fe_3O_4 films, such as molecular-beam epitaxy (MBE) [13,14], reactive ion beam deposition (IBD) [15,16] from an Fe target with Ar–O_2 mixture gas flow, pulsed laser deposition (PLD) from an oxide target [17,18], RF magnetron sputtering with an external inductively coupled RF source [19], and so on. The researches are focusing on the magnetic and structural properties for the single-crystalline Fe_3O_4. Here, Fe_3O_4 films were fabricated by DC magnetron reactive sputtering and then annealed in a vacuum system. During the sputtering, the magnetic field was usually applied to define the uniaxial magnetic anisotropy of the magnetic layer for using in spintronic devices [20,21]. At present researches, the magnetic field was seldom applied to fabricate the Fe_3O_4 film. So, the primary emphasis in our research was placed on applying an external magnetic field during the growth, and expected to use it in preparing spintronic devices.

2. Experiment

In this experiment, Fe_3O_4 thin films were fabricated by DC magnetron sputtering a Fe target in Ar + O_2 mixture gas. The argon and oxygen flow rates were fixed and only the sputtering powers were changed to find the exact condition for Fe_3O_4 to formation. For film preparation, LS500 automatic sputtering system was adopted. The base pressure in chamber before sputtering was better than 9×10^{-8} mbar, which can ensure the film quality, the working pressure was 1.3×10^{-3} mbar, and during the

*Corresponding author.
E-mail address: tangtang1227@163.com (X.-L. Tang).

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deposition the substrate temperature was kept at room temperature (RT). The optimized deposition condition, which means no other phase existed, for 150 nm-thick film was an oxygen flow rate of 1 sccm (sccm: standard cubic centimeter per minute), argon 100 sccm, sputtering power of 200 W, and then rapid annealing using an infrared lamp furnace system at 300 °C under vacuum [22]. In the film prepared procession, a Ta buffer layer, which was 20 nm, was used to avoid diffusing between the Fe_3O_4 film and the substrate [22].

In order to investigate the properties of Fe_3O_4 films growth with and without an external magnetic field, two series of 150 nm Fe_3O_4 films were fabricated as the optimized deposition condition. For the film fabricated with an external field, magnetic field with strength of 350 Oe was applied along the film plane. The saturation magnetization M_s was measured using a vibrating sample magnetometer (VSM). Their structures were characterized by X-ray diffraction (XRD) with a BEDE D1 XRD system, the composition and chemical states were analyzed by X-ray photoelectron spectroscopy (XPS), and the surface morphology was observed by SEM. In addition, the magnetoresistance (MR) properties were carried out with four-probe measurements.

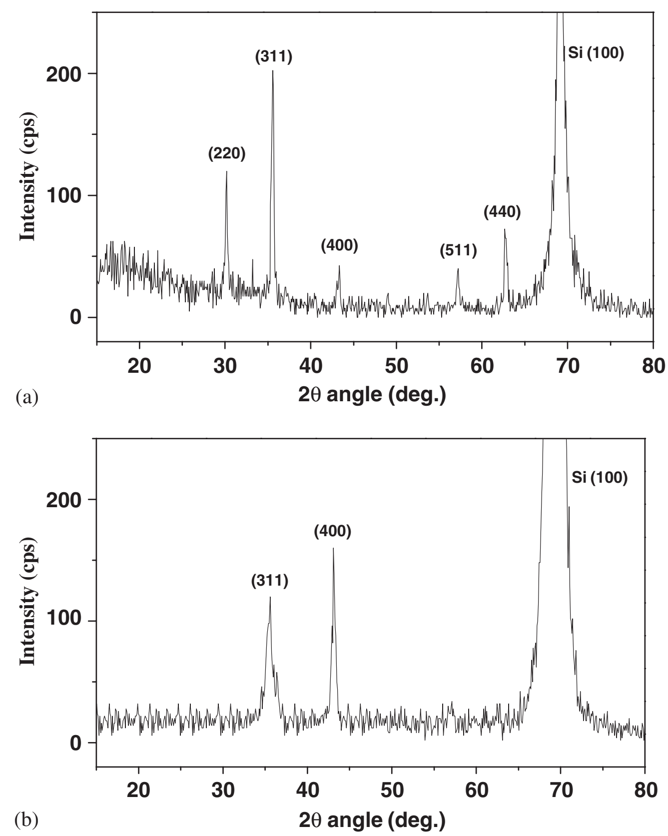


Fig. 1. XRD patterns of polycrystalline Fe_3O_4 films growth (a) without and (b) with an external magnetic field.

3. Result and discussion

3.1. Structure characteristics

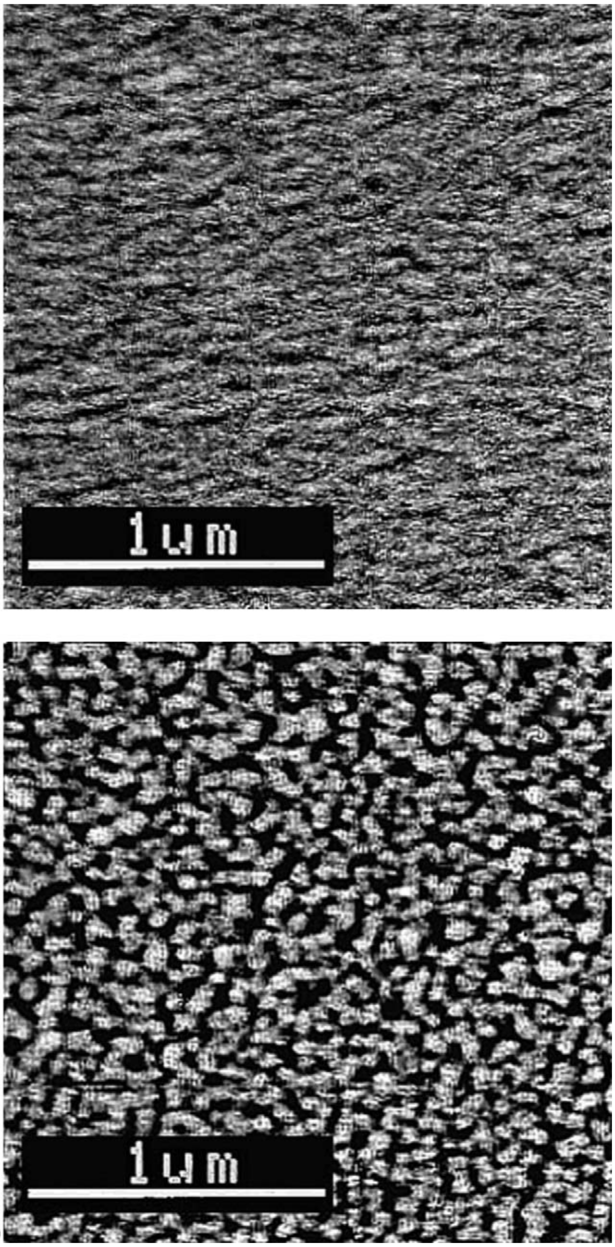
Fig. 1 shows the XRD patterns for the two series of films. The main peaks of the films were corresponding to the Fe_3O_4 cubic inverse spinel structures [19]. Due to the thin Ta buffer layer, its peaks does not appear in the XRD diagrams. For the film deposition without an external magnetic field, the obvious peaks corresponding to (220), (311), (400), (511), (440) orientations of Fe_3O_4 were observed, but when an external magnetic field of 350 Oe was applied along the film plane during the growth, the peaks corresponding to (200), (511), (440) disappeared. This suggests that Fe_3O_4 films grew with a preferred crystal direction of (311) and (400) when an external field was applied. Fig. 2 shows the surface morphology observed by SEM. The number of grains and crystal growth for films fabricated with an external field were improved, as compared to the sample fabricated without an external field.

In addition, due to the Fe_3O_4 has the nearly same structure and lattice parameter as maghemite (γ-Fe_2O_3), they cannot be distinguished from each other only by XRD, and the coexistence of small amounts of the γ-Fe_2O_3 can lower its MR values. So, in order to further confirm the chemical states presented in the film, XPS analyses were performed for the two series. Their patterns were the same, shown in Fig. 3. The XPS Fe 2*p* spectral peaks are broad and positioned around 711 and 724 eV, which are typical core level spectrum of Fe_3O_4. In addition, the absence of a satellite line at ~719 eV binding energy clearly reveals that γ-Fe_2O_3 does not exist in our samples [23].

3.2. Magnetic properties

Typical in-plane hysteresis loops of two series of the Fe_3O_4 films were measured using a VSM system at RT, as shown in Fig. 4.

It is evident from Fig. 4 that the magnetization measured at 14 kOe was 420 and 390 emu/cm^3 for the samples prepared with and without an external field at RT, respectively. The magnitude of the coercive fields, H_c, for the films prepared with and without an external field were almost the same (500 Oe), but the saturated magnetization fields for the two type films were different. The magnetization was saturated up to a field of 6 kOe when an external field of 350 Oe was applied to the sample during the prepared process. However, there was no sign of magnetization saturation in a field of up to 14 kOe for the film fabricating without an external field. It is well known that the magnetization of Fe_3O_4 thin film is not easy saturated because of the localized antiphase boundaries (APBs) in the Fe_3O_4 films [19]. According to our experiments, applying an external magnetic field to the sample during the growth is required for the efficient magnetic saturation of the film. In addition, we noted that the M_s values for our polycrystalline Fe_3O_4 samples are close to the value of



(a)

(b)

Fig. 2. Surface morphology of polycrystalline Fe_3O_4 films growth (a) without and (b) with an external magnetic field.

450 emu/cm^3 reported for epitaxial Fe_3O_4 thin film [24], and much higher than other groups reported for the polycrystalline Fe_3O_4 thin film [19]. The high M_s values were attributed to the Ta buffer layer, and Fig. 5 displays the hysteresis loops for the Ta-free and Ta-containing samples fabricated with an external magnetic field.

Fig. 6 shows the XRD pattern from a 150-nm Fe_3O_4 film deposited directly on a Si (100) substrate without a buffer layer. It is obvious that the intensities of the Fe_3O_4 reflections were greatly reduced compared with Fig. 1(a), most peaks disappeared, and the amorphous phase appeared. The amorphous phase is attributed to the deposition and annealing processes. During the deposition, iron and oxygen ion may take action at the Si surface and

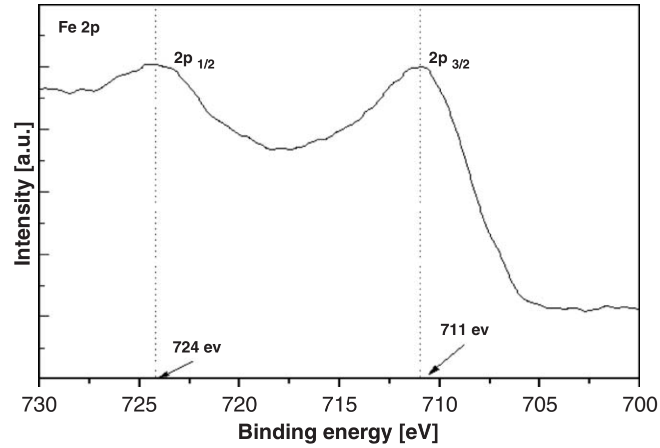


Fig. 3. Typical XPS measurement of Fe 2p core spectra of our Fe_3O_4 films.

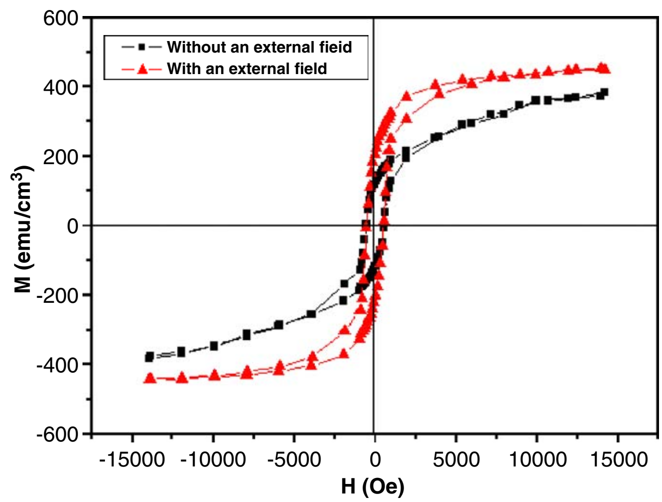


Fig. 4. Hysteresis curves of the polycrystalline Fe_3O_4 films growth with and without an external magnetic field.

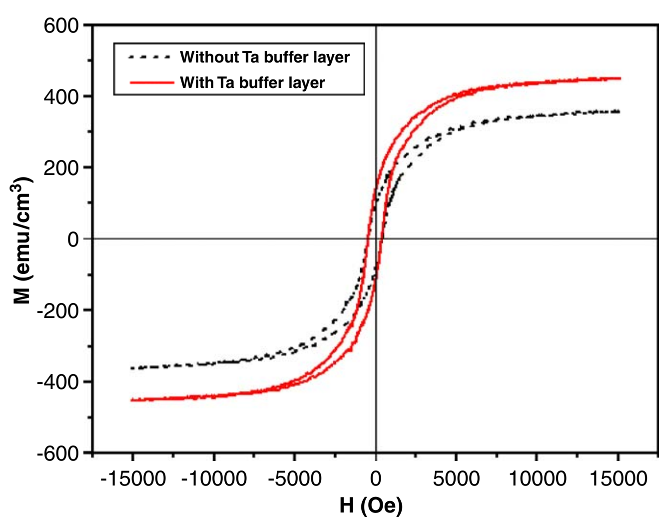


Fig. 5. Hysteresis curves of the polycrystalline Fe_3O_4 films growth with and without Ta buffer layer.

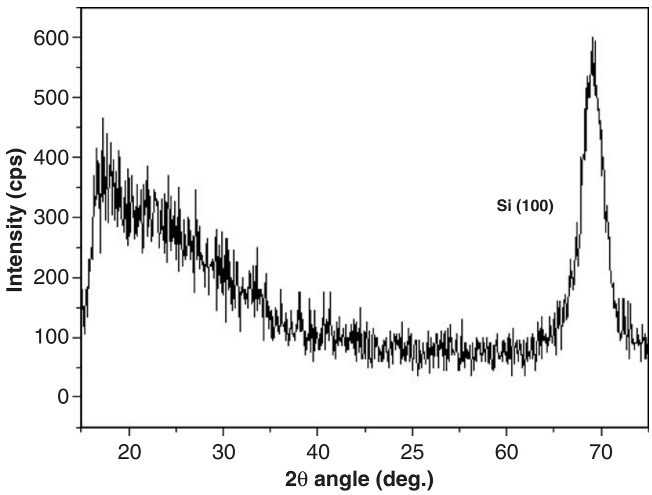


Fig. 6. XRD patterns of the 150 nm Fe_3O_4 films without a Ta buffer layer.

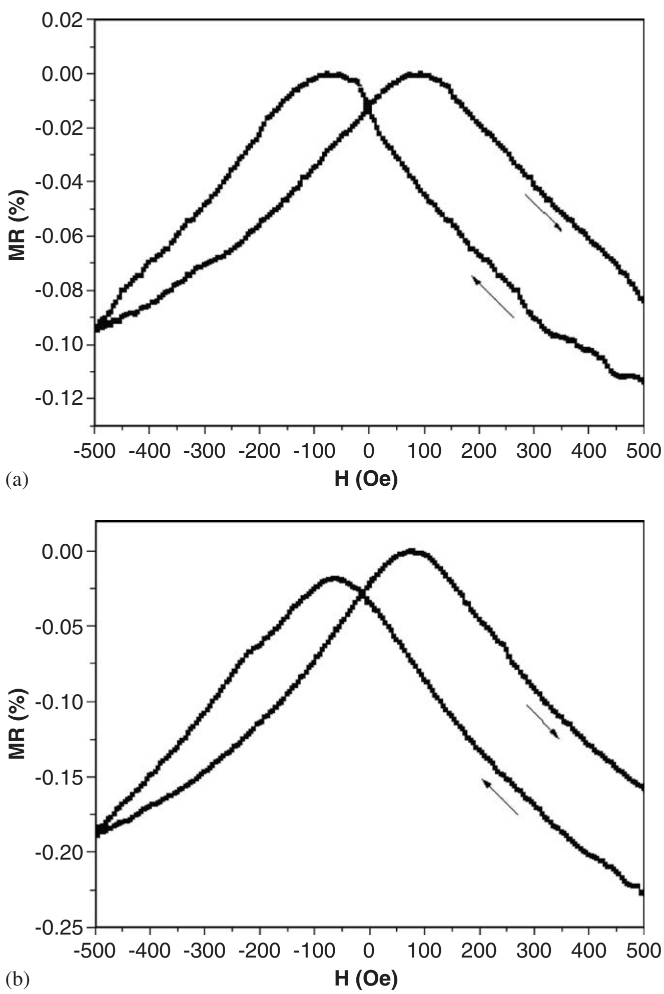


Fig. 7. Parallel MR measured at room temperature for samples prepared (a) without and (b) with an external magnetic field.

form amorphous phase just like the metal–SiO_2 system. In addition, the annealing process would accelerate the magnetic ions diffusing to the Si substrate and increase the amount of metal–SiO_2 compounds. So, the buffer layer prevented the oxidation and presented other phases at interfaces between the substrates and the Fe_3O_4 film and it also made the samples possess good structural properties.

3.3. *Magnetoresistance*

Fig. 7 shows the RT MR as a function of applied magnetic field for the sample prepared without and with an external field. Due to the limited measured field, we got only the MR curves (defined as $MR = (\rho_H - \rho_0)/\rho_0$) under 500 Oe.

The MR reaches -0.2% for the sample growth with an external field, which is two times larger than the sample growth without an external field. So, applying an external field during the deposition is also efficient to improve the MR value. Otherwise, the MR curves both present negative MR effects, which is one of the important properties for Fe_3O_4 films [25].

4. Conclusion

Half-metallic polycrystalline Fe_3O_4 thin films were successfully fabricated by DC magnetron reactive sputtering with excellent properties. Applying an external magnetic field to the sample during growth is efficient to modify the structure and to promote the MR values.

Acknowledgments

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