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Epitaxial Growth and Magnetic Properties of Fe_3O_4 Film on Si(111) Substrate

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Abstract

This study reports on the epitaxial Fe_3O_4 films on Si(111) substrate. To prevent the oxidization of the Si surface by introducing oxygen into chamber, γ-Al_2O_3 with spinel structure was inserted between Si substrate and Fe_3O_4 film. The Fe_3O_4 films were high-quality epitaxial films with two in-plane domains rotated by 180 degree. The magnetization properties showed that the Fe_3O_4 layer had in-plane magnetization.

1. Introduction

In the spintronics research fields, spinel ferrites are promising materials for spintronics applications because they have useful magnetic properties and high Curie temperature. In particular, Fe_3O_4 has been expected to be the spin source in the devices such as magnetic tunnel junctions[1] because Fe_3O_4 is a half metallic material[2]. However, direct fabrication of Fe_3O_4 on Si substrates is difficult because the surface of Si is oxidized by introducing oxygen into the chamber. Since Si is one of the universal and inexpensive substrates, it is important to fabricate spinel ferrites on Si substrate. In this study, to prevent the oxidization of the Si surface, we inserted the γ-Al_2O_3 with spinel structure as a buffer layer between Si and Fe_3O_4. We chose the γ-Al_2O_3 because it is possible to fabricate the thin film on Si substrate at ultra-high vacuum pressure($<10^{-6}$ Pa)[3] and same crystal structure. The heterostructure of Si substrate/ γ-Al_2O_3/ Fe_3O_4 might be a part of magnetic tunnel junctions or spin injection devises because Fe_3O_4 has half metallic property.

2. Experiments

The γ-Al_2O_3 and Fe_3O_4 were grown by the molecular beam epitaxy method. The sample structure was Si(111) /γ-Al_2O_3 2.4 nm /Fe_3O_4 50 nm /amorphous-Al_2O_3 2.0 nm. The γ-Al_2O_3, which is a nonmagnetic insulator, was used as buffer layer. The lattice constant is about 0.79 nm, that is just two third parts of Si[4]. The γ-Al_2O_3 film was formed at 900°C[5] and annealed at 300°C for 30 minutes. Then, the Fe_3O_4 film was formed by reactive deposition at 300°C in O_2 atmosphere of 4.0×10^{-4} Pa. The epitaxial growth and the surface structure were confirmed by reflection high energy electron diffraction (RHEED), atomic force microscope (AFM), X-ray diffraction (XRD) and transmission electron microscope (TEM). Cross-sectional samples for TEM were prepared using conventional mechanical polishing and dimpling techniques. The magnetic properties of Fe_3O_4 were measured by vibrating sample magnetometer (VSM).

3. Results and Discussion

The γ-Al_2O_3 film played a role of preventing the oxidization of Si surface and buffer layer for epitaxial growth. To confirm the epitaxial growth, we used RHEED. As shown in Fig. 1(a) and (b), The RHEED patterns of γ-Al_2O_3 and Fe_3O_4 were clear streak pattern. For surface morphology, the Root Mean Square (RMS) values of γ-Al_2O_3 and Fe_3O_4 were estimated 0.41 nm and 0.54 nm by AFM measurement. It means that both γ-Al_2O_3 and Fe_3O_4 were considered to be flat epitaxial films.

As shown in Fig. 2(a), the XRD diffraction pattern of θ-2θ measurement exhibits four peaks (18.3°, 37.2°, 57.2°, 79.4°), which is related to the diffraction of Fe_3O_4(111), (222), (333) and (444). It indicated that the Fe_3O_4 film was (111)-oriented without other orientations or phases. ϕ-scan measurements of Si(311) and Fe_3O_4(4-40) also exhibited that the Fe_3O_4 was epitaxial as shown in Fig. 2(b) . The six peaks of Fe_3O_4 (4-40) appeared as intervals of 60°, which indicates the presence of two in-plane Fe_3O_4 domains. It means that the domains were rotated by 180° and the epitaxial relationships are [11-2]Fe_3O_4(111) and [-1-12]Fe_3O_4(111) parallel to [11-2]Si(111).

As shown in Fig. 3, the cross-section TEM image and High-angle Annular Dark Field Scanning (HAADF) image of heterostructure taken along the Si[1−10] zone axis exhibits that the Fe atoms of Fe_3O_4 were aligned orderly and the

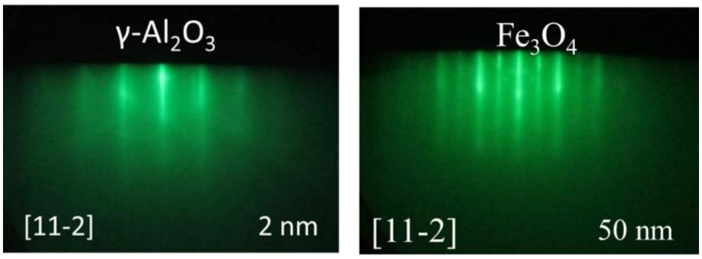

Fig. 1. RHEED patterns of (a) γ-Al_2O_3 and (b) Fe_3O_4

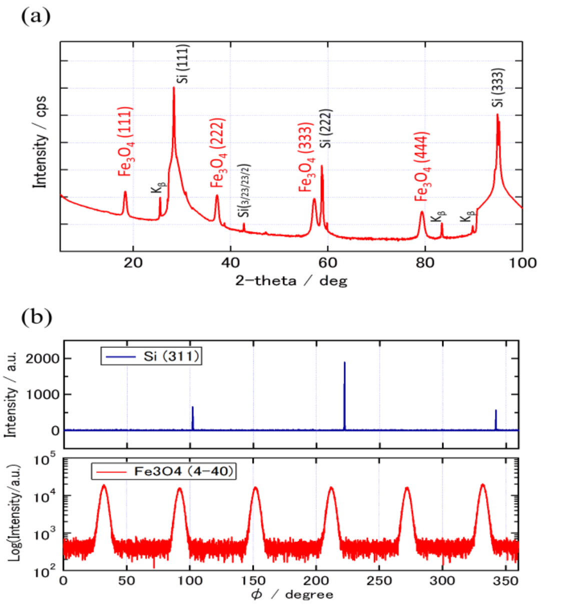

Fig. 2. X-ray diffraction patterns of (a) θ-2θ measurement (b) ϕ-scan measurement

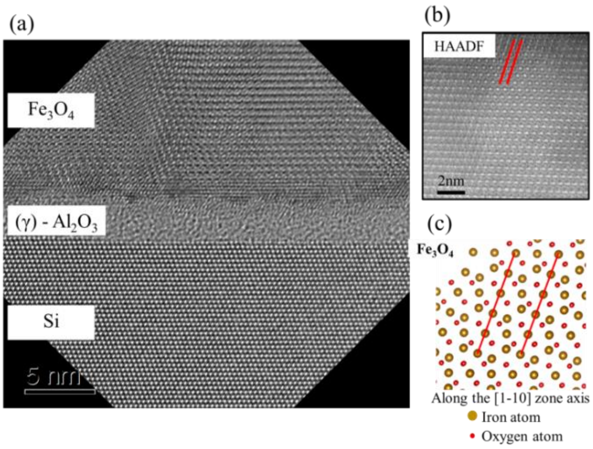

Fig. 3. (a)TEM images of Si(111)/ γ-Al_2O_3/Fe_3O_4 (b)HAADF image of Fe_3O_4 (c) The spinel structure lattice along the [1-10] zone axis

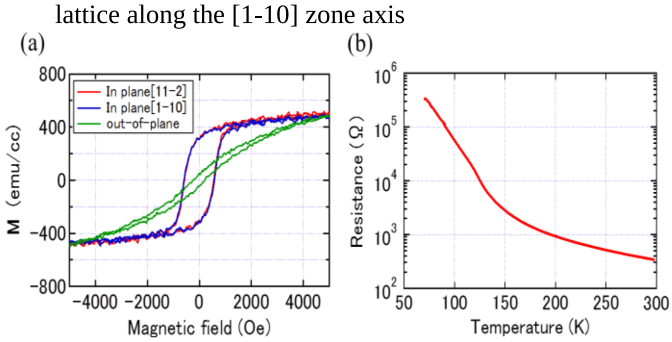

Fig. 4. (a) Hysteresis curves obtained from VSM measurements at RT (b) Dependence of resistance on temperature of Fe_3O_4 film

intervals between atoms were approximately corresponded to the spinel structure lattice parameter (5.14Å). In addition, γ-Al_2O_3 layer seemed to be amorphous which is inconsistent with RHEED observation. The reason is unclear, however the crystalline morphology might be changed during the growth of Fe_3O_4.

The interface between γ-Al_2O_3 and Fe_3O_4 was more unclear than the interface between Si substrate and γ-Al_2O_3. The lattice mismatch between γ-Al_2O_3 and Fe_3O_4 is 5.1%, which is larger than that between Si substrate and γ-Al_2O_3. Therefore, the Fe_3O_4 interface around the γ-Al_2O_3 is considered to be disordered.

The magnetization curves at RT for the Fe_3O_4 films are plotted in Fig. 4(a), where the directions of the magnetic field were in-plane[11-2], in-plane[1-10] and out-of-plane[111]. The saturation magnetization (Ms) was 580 emu/cc for all magnetic field directions. the remanent magnetization (Mr), the coercive field (Hc), and The remanent magnetization ratio (Mr/Ms) in the magnetic field along [11-2] were 280 emu/cc, 500 Oe, and 0.48, those for [1-10] were same, and those for [111] were 47 emu/cc, 225 Oe, and 0.08. In other words, The Fe_3O_4 layer had in-plane magnetization.

Fig. 4(b) shows that the dependence of the resistance on temperature for Fe_3O_4 film grown on the γ-Al_2O_3 buffer layer. The resistivity of the film at 300 K was 2.5×10^{-4} Ωcm. The curve shows an abrupt change at around 120 K, which corresponds to the Verwey transition[6][7] of Fe_3O_4. It means that the Fe_3O_4 film was considered to be high-quality.

3. Summary

We fabricated epitaxial Fe_3O_4 film on Si substrate by inserting γ-Al_2O_3 buffer layer. From the XRD measurement and TEM observation, Fe_3O_4 was (111)-oriented. The (111)-oriented Fe_3O_4 films grow with two possible in-plane domains. The magnetic hysteresis and the Verwey transition of Fe_3O_4 were clearly observed.

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