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Fabrication of $MgAl_2O_4$ thin films on ferromagnetic Heusler alloy Fe_2CrSi by reactive magnetron sputtering

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1. Introduction

In recent years, magnetic tunnel junctions (MTJs) using epitaxial MgO barrier have attracted great deal of interest because they show large tunnel magnetoresistance (TMR) ratios due to the highly spin-polarized electrons through the Δ_1 band of ferromagnetic electrode [1,2]. On the other hand, high spin-polarization can be also realized in MTJs using half-metal ferromagnets (HMFs), which have only one spin channel present at the Fermi level (E_F). Some of the Heusler alloys such as Co_2MnSi, Co_2MnGe [3] have been reported to be HMFs. According to the theoretical studies, coherent tunneling could occur and enhance TMR ratio by using $Co_2MnSi/MgO/Co_2MnSi$ MTJ [4]. However, the lattice mismatch between MgO and typical ferromagnetic materials including Heusler alloys is relatively large (around 4%). This large mismatch induces misfit dislocations (MDs) and antisite disorder at the interfaces of FM and MgO [5]. These defects drastically decrease the device performances. Therefore, tunnel barriers lattice matched with HMFs are necessary for high-performance spintronic devices.

Thus, we focused on magnesium aluminate spinel $MgAl_2O_4$ as lattice matched tunnel barrier with HMF Heusler alloy such as Fe_2CrSi. Lattice mismatch between $MgAl_2O_4$ (diagonal length $1/\sqrt{2} \times a_{MgAl2O4} = 0.5715$ nm) and Fe_2CrSi ($a = 0.5679$ nm [6]) is as low as +0.6%, which is considered to be small enough for coherent growth [7]. And, it has been reported that lattice matched $Fe/MgAl_2O_4/Fe$ MTJs showed the large bias voltage for one-half of the zero-bias TMR ratio (V_{half}) [8]. Band calculations suggested that Fe_2CrSi is robust against antisite disorder because it has half-metallic band structures with high density of states (DOS) at the E_F for the majority-spin band [9, 10]. Furthermore, its low saturation magnetization M_s ($1.98\mu_B$/f.u.) is useful for the spin-transfer switching, and Curie temperature (T_c = 630 K) is low enough for thermally assisted recording (but high against the room temperature) [9].

In this paper, we have fabricated $MgAl_2O_4$ thin films on MgO substrate and Fe_2CrSi thin films by reactive magnetron sputtering. We consider that $MgAl_2O_4$ thin film is very useful not only for the tunnel barrier and buffer layers of superconducting or ferroelectric films but also the practical use such as used in refractory ceramics and high pressure discharge lamps, but fabrication of $MgAl_2O_4$ epitaxial films by reactive magnetron sputtering is not reported so far.

2. Experiments

$MgAl_2O_4$ thin films were deposited on (001) MgO substrate by direct current (DC) reactive magnetron sputtering from a $MgAl_2$ target. All the deposition of $MgAl_2O_4$ thin films was performed in oxide mode with the sputtering current and bias voltage of 0.4 A and 200 V, respectively. The thicknesses of $MgAl_2O_4$ thin films on MgO were 90 nm, and the growth rate was 0.75 nm/min. The growth temperatures were varied from RT to 300ºC, and a typical total gas pressure was 30×10^{-3} Torr using mixture of O_2+Ar gases. The oxygen partial pressure (P_{O2}) was 1.2-5.0 $\times 10^{-3}$ Torr. The films were cooled down to RT under the deposition pressure. The multilayers consist of Fe_2VSi buffer (5 nm)/ Fe_2CrSi(50 nm)/$MgAl_2$(0, 0.2 nm)/$MgAl_2O_4$(45 nm) were prepared on (001) MgO substrate. The epitaxial Fe_2VSi and Fe_2CrSi Heusler alloy films were deposited by DC magnetron sputtering. The growth temperature of the Fe_2VSi and Fe_2CrSi were 650ºC and room temperature, respectively. Post-annealing was performed subsequently *in situ* after the deposition of Fe_2CrSi at 600ºC for 30 minutes. For structural characterization, a four-cycle and in-plane x-ray diffractometer equipped with a Cu Kα radiation source was used. Magnetic properties were measured by using vibrating sample magnetometer (VSM).

3. Results and discussion

$MgAl_2O_4$ thin films were fabricated on MgO substrate at the various temperatures and the oxygen partial pressure (P_{O2}) conditions. Figure 1(a) shows the growth results of $MgAl_2O_4$ thin films. Each symbol represents a growth condition where the lattice parameter was close to the bulk $MgAl_2O_4$ (closed circle), the XRD signals related to $MgAl_2O_4$ were clearly observed but the lattice parameter was smaller than the bulk value (open circle), and observed XRD signals were smaller and broader (triangle). Single phase $MgAl_2O_4$ films with good crystallinity were obtained under the P_{O2} condition of 5.0×10^{-3} Torr and in the growth temperature range from 200 to 350°C, as shown in Fig. 2(b). The (004) peaks of $MgAl_2O_4$ were clearly observed. Out-of-plane lattice parameter c of the $MgAl_2O_4$ film grown at 300°C was 0.8075 nm, which is very close to the bulk value of 0.8083 nm. Figure 1(c) shows the results of in-plane φ scan of the $MgAl_2O_4$ film grown at 300°C. XRD in-plane φ scans indicated $MgAl_2O_4$ film has a four-fold

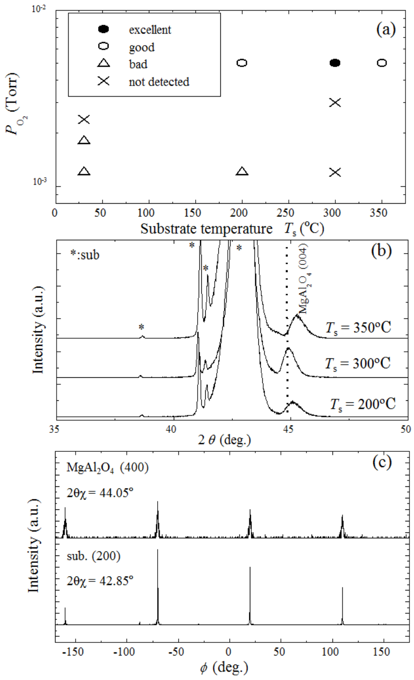

Fig. 1. (a) Film growth conditions and film crystallinity mapped in temperature versus P_{O2} diagram. (b) XRD patterns for the films grown at 200, 300 and 350ºC. (c) In-plane φ scans for the $MgAl_2O_4$ film grown at 350 ºC on MgO substrate.

symmetry, indicating epitaxial growth of $MgAl_2O_4$ film on MgO. In-plane lattice parameter a of $MgAl_2O_4$ deduced from (400) peak was 0.8223 nm, which is slightly large due to the strain induced by substrate. Thus, we have achieved the epitaxial growth of $MgAl_2O_4$ thin film on MgO by using reactive magnetron sputtering.

Next, we prepared the multilayered structures composed of MgO sub.//Fe_2VSi buffer(5 nm)/Fe_2CrSi(50 nm)/$MgAl_2$(0, 0.2 nm)/$MgAl_2O_4$(45 nm). XRD patterns of the multilayers with the thicknesses of $MgAl_2$ t_{MgAl2} = 0 and 0.2 nm are shown in Fig. 2. For the film of t_{MgAl2} = 0.2 nm, (002) and (004) peaks of Fe_2CrSi and (004) peak of $MgAl_2O_4$ were observed. The B2 ordering parameter S of Fe_2CrSi calculated from the integration of (002) and (004) was 0.90. On the other hand, for the film with t_{MgAl2} = 0 nm, (004) peak of $MgAl_2O_4$ was not observed. We consider that surface of the Fe_2CrSi was oxidized due to exposure under oxygen atmosphere during the deposition procedure of $MgAl_2O_4$. This means that $MgAl_2$ interlayer of 0.2 nm worked as protection layers for oxidization at the surface of Fe_2CrSi structure and made the growth of $MgAl_2O_4$ possible. We also investigated the in-plane configuration for the film with t_{MgAl2} = 0.2 nm, and observed a four-fold symmetry of the Fe_2CrSi (400) peaks (not shown here). However, we cannot observe the in-plane XRD patterns of $MgAl_2O_4$ because of the small mismatch between Fe_2CrSi and $MgAl_2O_4$. From the magnetization measurements at RT, we obtained the coercive field H_c of 11 Oe and M_s of 370 emu/cm^3, which is close to the calculated M_s of Fe_2CrSi with a $L2_1$ ordered structure (398 emu/cm^3) [9].

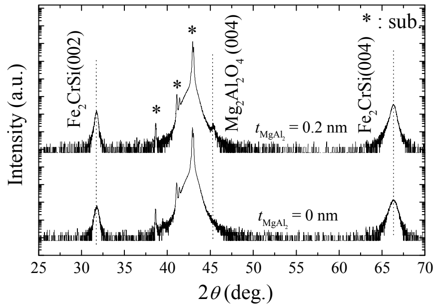

Fig. 2. XRD patterns of the multilayers with the thicknesses of t_{MgAl2} = 0, 0.2 nm.

3. Conclusions

In this present study, we have demonstrated the fabrication of $MgAl_2O_4$ epitaxial thin films on MgO substrate and Fe_2CrSi using a DC reactive magnetron sputtering technique. $MgAl_2O_4$ thin film grown on Fe_2CrSi was obtained by using the interlayer of $MgAl_2$ with a thickness of 0.2 nm before the deposition of $MgAl_2O_4$. After the deposition of $MgAl_2O_4$, the M_s of Fe_2CrSi film was kept close to the theoretical value with a $L2_1$ structure, which is expected to have half-metallic band structure. The present lattice-matched $Fe_2CrSi/MgAl_2O_4$ heterostructures are one of the promising candidates for spintronic tunnel devices.

Acknowledgments

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