Electrical properties of MgO insulating layers in spin-dependent tunneling junctions using Fe_3O_4

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We investigated the growth conditions and electrical properties of MgO epitaxial thin films, which have potential applications as insulating layers for spin-dependent tunneling devices where Fe_3O_4 serves as one of the magnetic electrodes. Our investigation showed that epitaxial MgO films with high crystalline quality can be successfully grown at temperatures as low as 473 K in oxygen pressures less than 1×10^{-5} Torr. This is a very important result because it indicates that the oxidation of the underlying Fe_3O_4 electrode is not a factor in fabrication of spin-dependent tunneling devices. We also examined the electron tunneling properties of Au/MgO/Fe_3O_4 junction with an ultrathin MgO layer prepared under the conditions described above and found excellent electron tunneling properties, as will be discussed. Barrier height and thickness estimated by curve fitting current density–voltage curves using the Simmons equation yielded barrier height and thicknesses of 0.9 eV and 2.5 nm, respectively. These values were consistent with those estimated by taking into account the reduction of the barrier height due to image forces. These results indicate that the MgO insulating layers grown under the restricted conditions have satisfactory electrical qualities required for spin tunneling devices. [S0021-8979(00)04421-2]

I. INTRODUCTION

Spinel ferrites with both ferrimagnetism and a broad range of electrical properties at room temperature are of great interest as materials for spin-dependent transport devices utilizing spin correlation between conduction electrons. One of the spinel ferrites, magnetite Fe_3O_4, has attracted much attention as a magnetic electrode material for tunneling junctions in which the magnetic electrode layers are separated by an insulating ultrathin layer.[1–3] That is because Fe_3O_4 is expected to be half-metallic and the half-metallic nature of the magnetic electrodes is believed to induce a large tunneling magnetoresistance in the junctions.[4,5] Thus, much current research related to Fe_3O_4 has focused on magnetotransport in grain boundary samples and in the tunneling junction structures.[6–9]

The magnetite contains 8 Fe^{3+} ions per unit cell in tetrahedral A sites and equal amounts of 8 Fe^{3+} and 8 Fe^{2+} ions in octahedral B sites. The stoichiometry, i.e., the ratio of Fe^{2+} to Fe^{3+} in octahedral sites, is sensitive to temperature and oxygen pressure in ambience because of high affinity of Fe^{2+} ions to oxygen, so that the deposited film, especially its surface, is readily oxidized when kept at high temperature and high oxygen pressure. This results in a serious problem in directly fabricating an insulating oxide layer on the Fe_3O_4 electrode layer: The surface of the Fe_3O_4 layer subjected to oxygen would be oxidized during deposition of the insulating oxide layer, which results in degradation of the half-metallicity of the Fe_3O_4 surface and hence a large reduction of the tunneling magnetoresistance effect. From the resistivity changes of the stoichiometric Fe_3O_4 thin films annealed at low oxygen pressures, we have recently found that Fe_3O_4 thin films are rapidly oxidized at temperatures higher than 573 K and/or at oxygen pressures higher than 1×10^{-4} Torr.[10] Therefore, growth of the insulating oxide layer with good electrical quality under the restricted conditions excluding the above ranges is strongly required, in order to avoid oxidation of the surface of the Fe_3O_4 layer and consequently to obtain a good spin-tunneling characteristic in a junction.

In this article, we describe the crystallographic quality of MgO epitaxial thin films grown under the restricted conditions, i.e., growth temperature <573 K and oxygen pressure $<1\times10^{-4}$ Torr, and discuss the electrical properties of the thin films through the current density–voltage (J–V) characteristics of trilayer junction structures in the form of Au (top electrode)/MgO (barrier)/Fe_3O_4 (bottom electrode). The MgO epitaxial film is promising as a barrier layer for the spin-dependent tunneling devices using the Fe_3O_4 magnetic electrodes because of a small lattice mismatch (0.3%) between MgO and Fe_3O_4.

II. EXPERIMENT

The MgO films for investigating the growth conditions were prepared on *c*-plane single crystalline sapphire substrates by pulsed laser deposition (PLD) using a Mg metal target. The sapphire substrate allows us to evaluate the structural qualities of the prepared MgO films through x-ray diffraction analysis, without interference between x-ray reflection peaks of the film and the substrate. The substrates were heated up to 773 K. A fourth harmonic (wavelength: 266 nm) of Nd: yttrium–aluminum–garnet (YAG) laser was employed as a laser source. The power density was 2 J/cm^2 on

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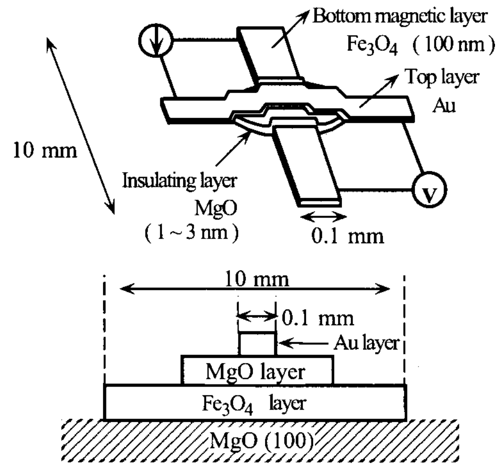

FIG. 1. Fabricated Au/MgO/Fe_3O_4 trilayer junction.

the target. The oxygen pressure $P(O_2)$ was held between 1×10^{-2} and 1×10^{-6} Torr during deposition by introducing oxygen gas through a precision leak valve. The deposition rates of the films were in the range of 0.3–0.5 nm/min.

The Au/MgO/Fe_3O_4 trilayer structures shown in Fig. 1 were fabricated on (100)-oriented MgO single crystalline substrates. Each layers were patterned through metal masks. The junction area was $1\times10^{-8}\,m^2$. The Fe_3O_4 bottom layers with about 100 nm thick were prepared at 573 K and $P(O_2)=1\times10^{-6}$ Torr by PLD using a ceramic target of Fe_3O_4, followed by the growth of MgO barrier layers.[10] The thickness of the MgO layers was adjusted by the deposition time, on the basis of the growth rate determined prior to these fabrications. The Au layers of the top electrode were deposited by conventional vacuum evaporation.

The structural properties of the prepared films were evaluated by x-ray diffractometry and reflection high-energy electron diffraction (RHEED). The surface of the Fe_3O_4 underlying layer was observed by atomic force microscopy (AFM). The $J-V$ characteristics of the trilayer junctions were measured by four-probe dc method in the range of room temperature to 170 K.

III. RESULTS AND DISCUSSION

Figure 2 shows x-ray diffraction diagrams of the MgO films deposited on c-plane sapphire substrates at various $P(O_2)$, where the growth temperature was fixed at 773 K. A strong (111) reflection of MgO appears at $P(O_2)\geqslant1\times10^{-5}$ Torr, indicating that the minimum $P(O_2)$ to oxidize the ablated Mg ions is around 1×10^{-5} Torr. It was confirmed from the RHEED measurements that the films grown at $P(O_2)\geqslant1\times10^{-5}$ Torr are single crystalline. Figure 3 shows the x-ray diffraction diagrams for the MgO films deposited at lower temperatures ($\leqslant$573 K) when $P(O_2)$ was fixed at 1×10^{-5} Torr. The (111) reflection of MgO was observed even for the film grown at room temperature, though it was rather broad. These results indicate that epitaxial thin films of MgO can be grown under the restricted conditions described above. However, as suggested from the shift of the x-ray peak positions to lower diffraction angles with growth temperature decreasing, the films grown at lower temperatures were more oxygen deficient. This is reflected on the electrical properties of the MgO films as will be seen below.

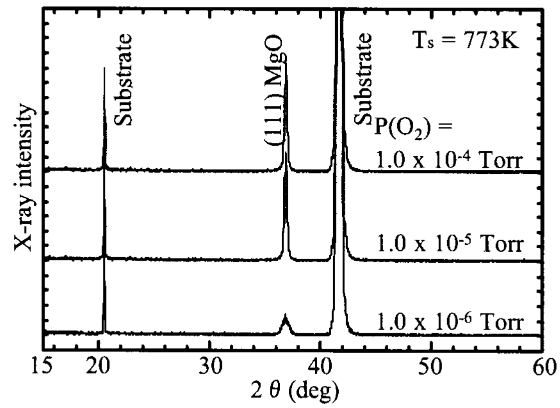

FIG. 2. X-ray diffraction diagrams of the MgO films grown on c-plane sapphire substrates at 773 K at various $P(O_2)$.

In order to evaluate the electrical quality of these MgO thin films grown under the restricted conditions, the $J-V$ characteristics of Au/MgO/Fe_3O_4 junctions were measured. The Fe_3O_4 thin films of the bottom electrode exhibited sharp and streaky RHEED images, along with Kikuchi lines and reflections from the higher-order Laue zones. The film surfaces were extremely smooth, having an average roughness as small as 0.1 nm, as shown in Fig. 4. This enables us to assess the intrinsic electrical properties of insulating ultrathin MgO layers free from pinholes, which cause local short circuit across the MgO layer. Figure 5 shows the $J-V$ curves at room temperature for the trilayer junctions with the 3-nm-thick MgO barrier layers deposited at (a) room temperature and (b) 473 K. Both curves were nonlinear, indicative of electron tunneling. The tunneling current density J transmitted through a rectangular potential barrier with height φ_0 and thickness s_0 is represented by the following equations, which were approximately derived for relatively low applied voltages $V(<\varphi_0/e)$ by Simmons[11]

$$J=\alpha(V+\gamma V^3), \tag{1}$$

$$\alpha=(3/2s_0)(e/h)^2(2m\varphi_0)^{1/2}\exp(-D\varphi_0^{1/2}), \tag{2}$$

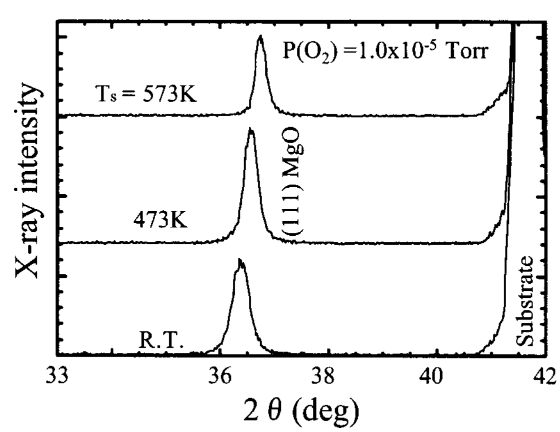

FIG. 3. X-ray diffraction diagrams of the MgO films grown on c-plane sapphire substrates at various temperatures.

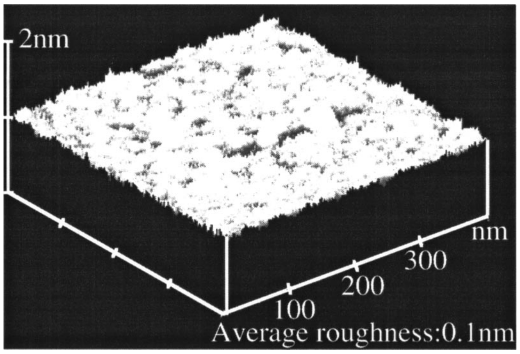

FIG. 4. AFM image for the Fe_3O_4 underlying layer grown on a MgO (100) substrate at 573 K and $P(O_2)=1\times10^{-6}$ Torr.

$$\gamma=(D^2e^2/96\varphi_0)-De^2\varphi_0^{-3/2}/32, \tag{3}$$

$$D=4\pi s_0(2m)^{1/2}/h, \tag{4}$$

where e is the charge of electron, m the mass of electron, and h Planck's constant. Equation (1) thoroughly fitted the measured curve (b) as shown in Fig. 5 (solid curve), while it did not fit the curve (a). The latter occurrence is presumably associated with the presence of leakage current through oxygen defects in the MgO layer, which is consistent with the above results of x-ray diffractometry. The leakage current is responsible for the steep rising of the J curve at low applied voltages as seen in (a). From the curve fit, the height and thickness of the barrier in the tunneling structure giving the curve (b) were determined to be 0.9 eV and 2.5 nm. However, these respective values were discrepant with the following expected values; the energy difference between the Fermi energy (4.0 eV) of Fe_3O_4 and the electron affinity (1.5 eV) of MgO, which corresponds to the barrier height, is 2.5 eV and the designed thickness of MgO insulating layer is 3 nm.[4] This discrepancy is removed by taking into account the reduction of substantial area of the potential barrier due to image forces. The height φ and thickness s of the barrier with the

$$\varphi=\varphi_0-[1.15\lambda s_0/(s_2-s_1)]\ln[s_2(s_0-s_1)/s_1(s_0-s_2)], \tag{5}$$

$$s=s_2-s_1, \tag{6}$$

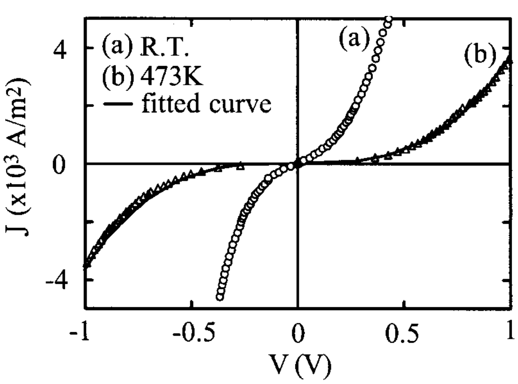

FIG. 5. $J-V$ curves measured at room temperature for the trilayer junctions with the 3-nm-thick MgO barrier layers deposited at (a) room temperature and (b) 473 K. The solid curve shows the fitted one using Eq. (1).

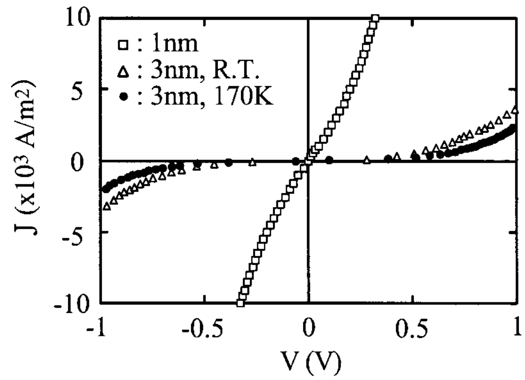

FIG. 6. $J-V$ curves measured at room temperature for the junctions with the MgO layers having different thicknesses. The curve at 170 K is shown for the 3 nm thickness, for comparison.

$$s_1=1.2\lambda s_0/\varphi_0, \tag{7}$$

$$s_2=s_0[1-9.2\lambda/(3\varphi_0+4\lambda-2eV)]+s_1, \tag{8}$$

$$\lambda=e^2\ln 2/8\pi\epsilon s_0, \tag{9}$$

where φ_0 and s_0 are the height and thickness of the barrier without the image force, respectively, and ϵ is the dielectric constant of vacuum. The values obtained for $V=1$ V using the above energy difference and the designed layer thickness were 1.0 eV in barrier height and 2.0 nm in barrier thickness, closer to the values estimated from the curve (b). The above results indicate that the MgO layer grown at 473 K and $P(O_2)=1\times10^{-5}$ Torr, which are within the restricted range of temperature and $P(O_2)$, has a satisfactory performance as a barrier layer for spin tunneling junctions.

Further confirmation of the high quality of the MgO films has been obtained from thickness and temperature dependencies of the $J-V$ characteristics. Figure 6 shows the $J-V$ curves of the junctions when the MgO layers with different thicknesses were grown at 473 K. The $J-V$ curve measured at 170 K was also shown for the junction with a 3-nm-thick MgO layer. Thinning the barrier layer resulted in the abrupt increase of electron tunneling probability, i.e., tunneling current, as is well known. The barrier height and thickness evaluated by the same manner as the above were 0.9 eV and 2.1 nm for the 1-nm-thick MgO layer. The evaluated barrier height was in agreement with that of the 3-nm-thick MgO layer, though the barrier thickness was considerably thicker than the designed one. The $J-V$ curve at 170 K was similar to that at room temperature, which ensures that strongly temperature-dependent current such as leakage current is negligibly low. These results emphasize the high quality of the MgO ultrathin layer grown at 473 K.

IV. CONCLUSION

We have epitaxially grown MgO thin films on sapphire substrates and MgO substrates with Fe_3O_4 underlying layers by PLD, and investigated the structural and electrical properties of the thin films. It was found that MgO thin films with high crystalline quality are grown at 473 K and $P(O_2)=1\times10^{-5}$ Torr, at which no degradation of the Fe_3O_4 underlying layer occurs. The $Au/MgO/Fe_3O_4$ junctions with the

MgO ultrathin layers prepared under the above restricted conditions exhibited the nonlinear $J-V$ curves that are characteristic of electron tunneling. The barrier height and thickness estimated from the curve fitting were 0.9 eV and 2.5 nm for the 3-nm-thick MgO film, respectively. This is consistent with the values roughly estimated by taking into account the reduction of the barrier height due to image forces. These results indicate that MgO insulating layers with high electrical quality for spin-dependent tunneling devices can be successfully grown on Fe_3O_4 underlying layers without making the surface of Fe_3O_4 layer oxidized.

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