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Effects of oxygen gas pressure on properties of iron oxide films grown by pulsed laser deposition

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ABSTRACT

Iron oxide films were grown on sapphire substrates by pulsed laser deposition at oxygen gas pressures between 1×10^{-5} and 1×10^{-1} Pa with a substrate temperature of 600 °C. Atomic force microscope, X-ray diffraction, Raman spectroscopy, X-ray absorption fine structure, and vibrational sample magnetometer analysis revealed that surface morphology and crystal structure of the iron oxide films strongly depend on the oxygen gas pressure during the growth and the optimum oxygen gas pressure range is very narrow around 1×10^{-3} Pa for obtaining single phase magnetite films with high crystal quality.

1. Introduction

Magnetite has attracted enormous attention because of its half metallic ferromagnetic nature, high Curie temperature, and the presence of a metal–insulator transition [1–3]. These properties make it a promising material for applications in spin electronic devices such as magnetic tunnel junctions for magnetic random access memory [4]. Many growth methods such as electron beam deposition [4], molecular beam epitaxy [5–7], and sputtering [8–11], have been used to fabricate magnetite films. The magnetite film on sapphire system is an important candidate for heteroepitaxial magnetic tunneling junction structures which exhibit giant tunnel magnetroresistance [12]. Pulsed laser deposition (PLD) is a promising technique for preparing thin films due to its advantages which include stoichiometric transfer, growth from an energetic beam, reactive deposition, and inherent simplicity for the growth of multilayered structures [13]. Especially, for growing nanometer order thin films, PLD is one of the most advanced techniques because the growth can be controlled precisely. In PLD process for growing iron oxide films, oxygen gas pressure is one of the most important parameters affecting on thin film properties. Parames et al. [14] tried to grow magnetite thin films on sapphire substrates by PLD in reactive atmospheres of oxygen and argon, at working pressure of 8×10^{-2} Pa. They have investigated the microstructure and magnetic properties of the obtained magnetite films and have reported the saturation magnetization of the magnetite film is about 315 emu/cm^3, which is much smaller than the value for bulk magnetite. In this article, we present on the effects of oxygen pressure on the properties of the iron oxide films in PLD because a deeper knowledge on the growth of the magnetite thin films with high quality is essential for device applications. We have succeeded in fabrication of magnetite film with saturation magnetic moment close to the value of bulk magnetite.

2. Experimental

Iron oxide films were grown on *c*-plane sapphire substrates by pulsed laser deposition in atmosphere of oxygen. The sapphire substrates were cleaned ultrasonically in organic solvents, chemically etched in a hot H_3PO_4:H_2SO_4 (1:3) solution, rinsed in deionized water, and blown dry in nitrogen gas before they were introduced into the growth chamber. Pulsed KrF excimer laser at a wavelength of 248 nm with a repetition rate of 2 Hz and a energy density of 2 J/cm^2 at the target surface was used to ablate magnetite target (99.9%). The substrate to target distance was 50 mm. High purity oxygen gas were introduced through mass flow controllers after the growth chamber was evacuated below 1×10^{-5} Pa using a turbomolecular pump. The target was rotated during the growth to avoid crater formation. The substrate temperature was kept at 600 °C while the oxygen gas pressure was controlled at a given value between 1×10^{-5} and 1×10^{-1} Pa. After the growth, thickness of the iron oxide films was determined by a surface step profile analyzer. The thickness of these films varies from 44.5 to 80.4 nm. Since the thickness of the film

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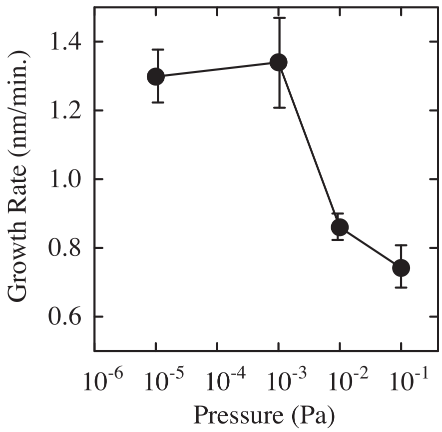

Fig. 1. Dependence of the growth rate of the iron oxide films on oxygen gas pressure.

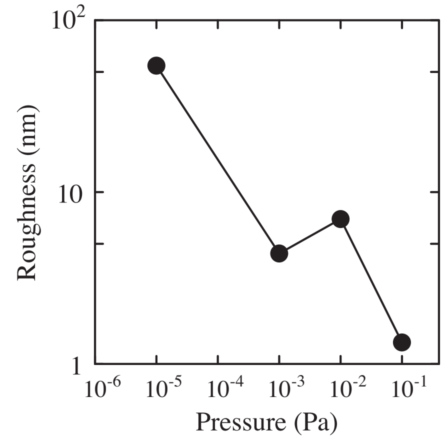

Fig. 3. Dependence of the surface roughness of the iron oxide on oxygen gas pressure.

grown at a given oxygen gas pressure increased linearly with increasing the growth time, we determined growth rate simply dividing film thickness by growth time. The crystallographic structure of the iron oxide films was analyzed by X-ray diffraction (XRD) using Kα emission line of copper. Raman measurements were performed on a micro-Raman system equipped with a classic charge-coupled device detector. The Raman scattering was recorded in the backscattering geometry of the $z(x,-)\bar{z}$ configuration using an Ar ion laser at 488 nm. The surface morphology and roughness were studied by atomic force microscope (AFM) on $10 \times 10\ \mu m^2$ areas under ambient conditions. X-ray absorption fine structure (XAFS) experiments were carried out at beam line BL11 of Saga Light Source with a Si (111) double crystal monochromator using synchrotron radiation. The XAFS spectra were collected by recording the conversion electron yield at room temperature. Vibrational sample magnetometer (VSM) was used to investigate the magnetic properties of the iron oxide films.

3. Results and discussion

Fig. 1 shows the dependence of the growth rate of the iron oxide films on oxygen gas pressure. The growth rate remains almost constant when the pressure is increased to 1×10^{-3} Pa, then decreases with further increasing pressure. The largest growth rate at the pressure of 1×10^{-3} Pa is observed to be 1.34 nm/min, suggesting that the thickness of the grown films can be controlled precisely in our PLD growth system.

Fig. 2 presents AFM images of the iron oxide films grown at various oxygen gas pressures. The root mean square (RMS) surface

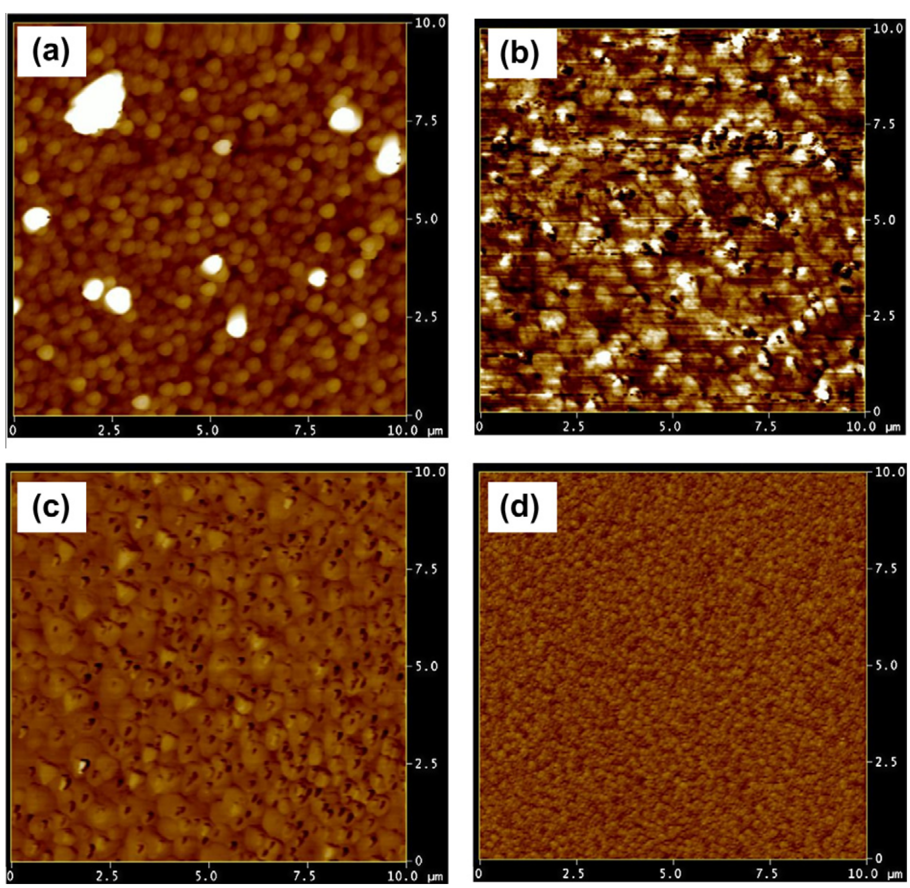

Fig. 2. AFM images of the iron oxide films grown at different oxygen gas pressures: (a) 1×10^{-5}, (b) 1×10^{-3}, (c) 1×10^{-2} and (d) 1×10^{-2} Pa.