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Effect of oxygen pressure on electrical transport properties for (110) oriented La_{2/3}Sr_{1/3}MnO₃ films directly deposited on silicon

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Abstract: $La_{2/3}Sr_{1/3}MnO_3$ films with (110) preferred orientation were deposited on Si (100) substrate without any buffer layer by pulsed laser deposition technique. Effect of oxygen pressure on orientation, surface morphology, and electrical transport properties were investigated. The film deposited at 10 Pa presented (110) preferred orientation with the best crystalline quality, the largest grain size, and the smallest roughness. The (110) oriented film presented higher metal-insulator transition temperature, and the lower resistivity than that of the samples without preferred orientation.

Keywords: colossal magnetoresistive film; LSMO; pulsed laser deposition; electrical transport; rare earths

Colossal magnetoresistance (CMR) in the doped manganese oxides has attracted much attention due to its potential applications in spintronic devices^[1-3], such as magnetic field sensors, magnetic memory devices, hard disk read heads, etc.^[4] Among these oxides, La_{2/3}Sr_{1/3}MnO₃ (LSMO) is a promising material for applications due to its nature half-metallic ferromagnetic properties and high Curie temperature $(T_{\rm C}>300 \text{ K})^{[1]}$. Furthermore, LSMO should be fabricated in thin film form to satisfy the request of the above mentioned applications. Recently, it has been reported that (110) oriented film of La_{1-x}Ae_xMnO₃ (Ae=Ca, Sr) presents enhanced magnetic properties compared to (100) oriented films^[5,6], such as an enhanced saturated magnetization and higher Curie temperature. Additionally, the thickness of interfacial magnetic dead layer formed at the (110) oriented LSMO/STO interface is suppressed compared to the (100) oriented one^[7-9]. Thus, it is expected that the per-</sup> formance of magnetic device can be improved by using a (110) oriented LSMO thin film due to its enhanced magnetic properties.

The LSMO films have been successfully deposited on some substrates, such as SrTiO₃, LaAlO₃ and MgO single crystal^[9–12], and ZnO^[13] nanorod arrays, etc. In particular, integrating LSMO onto Si, the essential material of the semiconductor industry, is crucial in maximizing their potential use^[14–17]. It is generally accepted that a

buffer layer is usually needed for most LSMO films deposited on Si with a preferred orientation, which makes the whole technique much more complicated. In this study, by using a pulsed laser deposition system, (110) preferred orientation $La_{2/3}Sr_{1/3}MnO_3$ films were successfully deposited on Si (100) substrate without any buffer layer. The effects of oxygen pressure on the orientation, surface morphology were investigated. Furthermore, electrical transport property was also studied because it can indirectly reflect the magnetic properties of these films.

1 Experimental

A series of La_{2/3}Sr_{1/3}MnO₃ films of about 50 nm were directly deposited on the Si (100) substrate by using pulsed laser deposition. The La_{2/3}Sr_{1/3}MnO₃ ceramic target was ablated by a KrF excimer laser (λ =248 nm) with energy density of 1.5 J/cm² and a laser repetition rate of 5 Hz. All of the films were deposited at 700 °C, and the distance between the target and substrate was around 50 mm. The Si (100) substrates were cleaned by soaking them in an ultrasonic bath in toluene, acetone, and ethanol for 15 min in turn. Then, a 25% hydrofluoric acid (HF) solution was used to remove the native SiO₂ of Si substrate. The chamber was evacuated to 2×10⁻⁴ Pa, and then an amorphous LSMO film was deposited at room

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temperature before initial deposition, which can avoid the formation of SiO_2 layer at high temperature.

The structure of the films was examined by X-ray diffraction (Bruker D8 advance, XRD) using a Cu K α radiation with wavelength of 0.15406 nm. The thickness was measured by a Seimitzu Surfcom 480A profiler. In order to correctly evaluate the growth rate of the LSMO film, scanning electron microscopy (SEM, Hitachi S4800) was also used to detect the cross section of the film. The surface morphology was studied by scanning electron microscopy (SEM, Hitachi S4800) and atomic force microscopy (AFM, Solver P47 Pro). The atomic force microscopy was also used to examine the surface roughness. Resistivity was measured by the four-probe method using the automatic measurement system assembled by Agilent E5273 and Lakeshore 340.

2 Results and discussion

Fig. 1(a) illustrates the XRD patterns of the films deposited at 1–50 Pa. All diffraction peaks are indexed to perovskite LSMO phase except for the film deposited at 1 Pa. The amorphous film deposited at 1 Pa might be related to the bad oxygen deficiency at a low pressure. It is worth noting that the film grown at 10 Pa shows the highest integrated intensity of (110) reflection peak. Above 10 Pa, only (110) reflection peak can be observed, and the integrated intensity decreases from 10 to 50 Pa, whereas full width at half maximum (FWHM) of the (110) peak increases. It means that crystallinity of the films gradually decreases with the increase of oxygen pressure.

There are two factors contributing to this phenomenon. Firstly, it relates to the lattice mismatch, which can

strongly affect the film orientation and crystallinity^[18-22]. The lattice mismatches between La_{2/3}Sr_{1/3}MnO₃ film and Si (100) substrate can be evaluated via the definition of $|a_{\rm f}-a_{\rm s}|/a_{\rm s}\times 100\%$, where $a_{\rm f}$ and $a_{\rm s}$ denote the in-plane lattice constants of the deposited film and substrate, respectively. The lattice constant of Si is 0.543 nm, and the pseudocubic lattice constant of La_{2/3}Sr_{1/3}MnO₃ is 0.388 nm. As a result, the lattice mismatch of the (100) plane between La_{2/3}Sr_{1/3}MnO₃ and Si is 7.2%. However, the interatomic distances along the <110> and <100> crystal orientation are 0.546 and 0.579 nm in the (110) plane of La_{2/3}Sr_{1/3}MnO₃, respectively, and corresponding lattice mismatch is 0.5% and 6.5% between the $La_{2/3}Sr_{1/3}MnO_3$ <110> and <100> crystal orientation and Si (100) plane. Accordingly, the (110) plane growth is easier than (100) plane when LSMO deposits on Si. Fig. 1(b) shows that the reflection angle of (110) peak decreases with increasing oxygen pressure, which suggests the increase of LSMO lattice constant. It can be attributed to the lattice expansion which derives the increase of oxygen content in the film^[23]. Consequently, we infer that the lattice mismatch between (110) oriented LSMO film and Si (100) substrate can reach minimum at 10 Pa, which leads to a best crystalline quality.

Secondly, at higher ambient oxygen pressure, the ablated species lost their initial energy continuously because of frequent collisions with oxygen atoms before they arrive at the substrate^[19,21,23]. Therefore, with increasing pressure, the particles ablated from the $La_{2/3}Sr_{1/3}MnO_3$ target do not have sufficient energy to undergo the surface diffusion, which results in the deterioration of crystallinity^[21]. Therefore, the highly crystalline (110) oriented LSMO film can be prepared by modulating oxygen pressure during deposition.



Fig. 1 XRD patterns of La_{2/3}Sr_{1/3}MnO₃ films directly deposited on Si substrate at various oxygen pressures (a) and oxygen pressure dependence of position and FWHM for (110) diffraction peak (b)

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