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Colossal Magnetoresistance in $\text{La}_{1-x}\text{Pb}_x\text{MnO}_3$ Thin Films Prepared by Pulsed Laser Deposition

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Abstract. $\text{La}_{1-x}\text{Pb}_x\text{MnO}_3$ thin films were prepared by off-axis pulsed laser deposition from stoichiometric targets on SrTiO_3 and LaAlO_3 substrates using KrF excimer laser radiation (248nm). Thin film deposition was performed at substrate temperatures between 650°C and 850°C and an oxygen pressure of 0.4mbar. Thin films prepared at 750°C were single phase and (001)-oriented. Epitaxial film growth was confirmed by x-ray pole figures and RHEED. Magnetic properties were studied by SQUID magnetometry. Ferromagnetic behavior with Curie temperatures between 200°C and 300°C was observed to be dependent on deposition parameters. Magnetoresistance was examined by standard 4-point measurements in fields up to 8T. As an example, a film with $T_c = 193\text{K}$ shows a resistance change $((\rho(0) - \rho(H)) / \rho(0))$ of about 25% at $T = 300\text{K}$. For $T = 220\text{K}$ and 180 K the changes are 70%.

1. INTRODUCTION

In the past years the giant magnetoresistance (GMR) behavior in metallic multilayers received much attention due to possible industrial applications in magnetic sensors. Recently, large GMR effects called colossal magnetoresistance (CMR) effects were reported for perovskite oxides such as $\text{La}_x\text{A}_{1-x}\text{MnO}_3$ (A : Sr, Ca, Ba, etc.). The CMR ratios $((\rho(0) - \rho(H)) / \rho(H))$ so far reported ranged up to $1 \times 10^5\%$ at 77K for La-Ca-Mn-O films [1] and $5 \times 10^5\%$ at 88K for Pr-Sr-Ca-Mn-O [2]. At or close to room temperature, which is the necessary operating temperature for technical applications, the CMR effects are much smaller : 150 % at 300K and 7T for La-Ba-Mn-O thin films [3] or 400% in an annealed La-Ca-Mn-O film at 280K and 6T. Another interesting system which shows high CMR ratios in bulk samples of up to 560% at 320K and 6T is La-Pb-Mn-O [4-6]. In this work we report on the preparation of La-Pb-Mn-O thin films and their characterization.

2. EXPERIMENTAL

Bulk targets of $\text{La}_x\text{Pb}_{1-x}\text{MnO}_3$, where $x=0.2$, were prepared from a stoichiometric mixture of PbO_2 , La_2O_3 and MnCO_3 by standard calcination (800 °C, 24 h) and sintering (1200 °C, 24h) procedures. The resulting targets were single phase with a density of 74% of the theoretical density. A KrF (248nm) excimer laser was used to deposit the films on (100) SrTiO_3 (STO) and LaAlO_3 substrates using an energy density of 2 J/cm² at the target. The deposition was carried out in a pure O_2 atmosphere of 0.4 mbar using the off-axis deposition geometry [7]. The substrate temperature during thin film deposition ranged between 650 °C and 850 °C for a substrate to target distance of 6 cm. After the deposition process the films cooled down to room temperature in 400 mbar O_2 . The typical film thickness was about 200 nm. X-ray diffraction and RHEED have been used for the structural characterization of the films. Magnetization measurements were performed using a commercial SQUID magnetometer. The magnetoresistance measurements were made using a conventional four-point technique in a temperature range of 4K to 300K and magnetic fields up to 8T. The CMR ratio is defined as $\rho(H) - \rho(0) / \rho(0)$.

3. RESULTS AND DISCUSSION

X-ray diffraction indicate that the deposited films have the perovskite-type cubic structure with a lattice parameter of $a = 3.895 \text{ \AA}$. They were single phase with a strong (001) orientation. Epitaxial film growth on LaAlO_3 was proven by X-ray pole figures and RHEED.

Figure 1 gives the temperature dependence of the magnetization of a thin film deposited at a substrate temperature of 750 °C, measured in an applied field of 0.1 T. The Curie point $T_c = 193$ K is defined as the point where dM/dT reaches the maximum value. A linear extrapolation of the high temperature susceptibility $1/\chi$ gives the paramagnetic Curie point $\Theta_p = 205$ K. The reduction of T_c compared to bulk samples ($T_c = 320$ K [4]) is possibly due to film oxygen deficiency which changes the doping level and therefore the mixed Mn^{3+}/Mn^{4+} ratio determining the ferromagnetic behavior, as stated for La-Ba-Mn-O thin films [8]. This oxygen deficiency is partly caused by the off-axis deposition geometry, as it was shown for YBaCuO thin films [7]. The Curie point of films deposited at different temperatures shows a maximum of $T_c = 198$ K at a substrate temperature of 700 °C. The reduced T_c at high substrate temperatures (> 800 °C) is possibly due to loss of Pb during the thin film deposition process.

Figure 2 shows the magnetoresistance effect at temperatures around the Curie temperature and at room temperature with the current parallel to the magnetic field. The CMR ratios at 8 T are about 25% at room temperature and increased to 70% at 180 K/220 K. The change in the curvature of the magnetoresistance below and above the Curie temperature is similar to that observed in La-Pb-Ca-Mn-O single crystals [9]. Therefore the low-field magnetoresistance effect is extremely different below and above the Curie temperature. At 0.1 T and 20 K below T_c the magnetoresistance ratio is 3%, but only 0.12% 20 K above T_c . This behavior shows, that it is more useful for potential applications to increase the Curie temperature of the thin films to room temperature than optimizing the high field CMR ratio at low temperatures.

In summary, epitaxial La-Pb-Mn-O thin films were prepared by off-axis pulsed laser deposition on $SrTiO_3$ and $LaAlO_3$ substrates. The films showed reduced Curie temperatures compared to bulk samples of the same composition due to oxygen deficiency. The magnetoresistance ratio near the Curie temperature shows values up to 70% at 8 T. At room temperature the films show a magnetoresistance ratio of about 25%.

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Figure 1 : Magnetic moment vs. temperature dependence of a LaPbMnO thin film deposited at 750°C. The inset shows the linear extrapolation of the high-temperature susceptibility $1/\chi$

Figure 2 : Magnetic field dependence of the resistance at 180K, 220K and 300K