Pulsed Laser Deposition of epitaxial SrTiO₃ films: Growth, structure and functional properties

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Abstract

Langmuir ion probe and in situ RHEED were applied to study the effects of low oxygen pressure on SrTiO₃ (STO) film growth by Pulsed Laser Deposition (PLD). Contributions of different oxygen fluxes were analysed and parameters of STO epitaxial film growth were evaluated using physical model of adiabatic expansion of the ablation products and its interaction with ambient gas. Film surface undergoes reconstruction at growing temperatures >600 °C indicating complete or partial relaxation of top layer without changing growth mechanism of smooth multilayered film. All films have a tetrahedral lattice distortion in the direction of growth that varying with deposition temperature and oxygen pressure. STO lattice distortion is the relevant factor in determining both agility and dielectric loss for tuneable microwave devices. Annealing in oxygen at 1100 °C improves significantly functional properties of STO films, but only the layers deposited under the pressure lower than 10^{-3} Pa possesses low dielectric losses in combination with high agility.

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1. Introduction

Progress in modern nanotechnology is concerned with the development of new deposition methods and also with the improvement of precision and reproducibility of available techniques. Necessary thicknesses, structure or functional properties of atomic layers and films are usually obtained by monitoring of growth conditions, for this purpose correlation curves, empirical relationships or proper physical models are used. Nevertheless the precision and reproducibility of widely used Pulsed Laser Deposition (PLD) remained insufficient for some applications of thin oxide films. Despite PLD experimental simplicity, a deposition from laser plasma in low-pressure gas, for example, in oxygen, is quite a complex process involving important changes in plasma flux during propagation from target to substrate. Hence, as a rule, the actual conditions of films nucleation and growth can be estimated only indirectly from the obtained PLD results. In spite of the fact that several physical models and theoretical studies of PLD process have

been proposed almost a decade ago [1–3], nevertheless, the experimental data in these publications were more likely illustrative, and during the last years these publications practically were not applied for in-depth analysis of new PLD data. The aim of this study is quantitative evaluation of STO film deposition processes in low-pressure oxygen and estimation of accordance between PLD theoretical models and experimental results of oxides target sputtering. These studies relate to the optimisation of functional properties STO ferroelectric films, for example, for application in fast agile microwave filters.

Experimental studies of ablation fluxes from SrTiO₃ target. First direct and accurate measurements of film deposition rates were fulfilled using PLD technique supplemented with in situ RHEED [3]. STO films with a flat surface were grown in a high vacuum or at low gas pressure P_g, where an interaction between the evaporated atoms and ambient gas is negligible. After this, the total number of evaporated atoms N_0 and effective deposition rate $i(t) = h(L)/t$ can be simply calculated from the film thickness h obtained at a given distance L:

$$N_0 = \gamma \cdot n_L L^2 \gamma \cdot h(L) = \gamma \cdot \rho \frac{N_A}{M} L^2 \cdot h(L), \quad (1)$$

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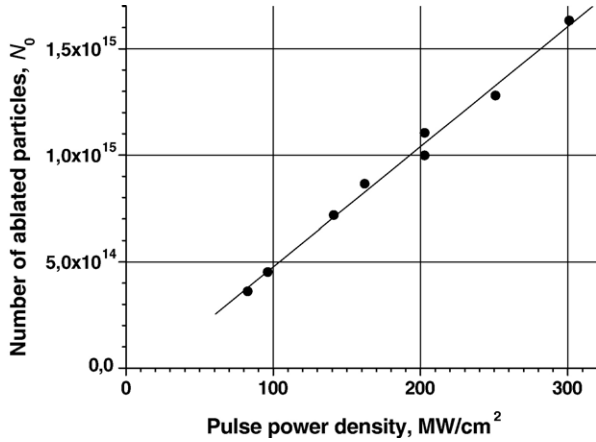 *E-mail address:* anatole.khodan@mail.ru (A.N. Khodan).

Fig. 1. The number of ablated particles with a mean mass $m_a=(Sr, Ti)$ as a function of laser pulse power density on the STO target in $1.3 10^{-3}$ Pa of oxygen.

where: $\gamma-$ is a geometrical factor; $\gamma \approx 1$ as the laser spot diameter on target $r_0 \ll L$; n_L — density of deposited film; typical n_L value for the solids is $\approx 5 \times 10^{22}$ cm^{-3}. STO films

deposition conditions vary about: $N_0=10^{16}-10^{17}$, $h=0.1$ nm/pulse and $L=3-10$ cm.

The obtained correlation between laser pulse power density and the total number of the evaporated atoms $N_0=\gamma\, n_L L^2\, h(L)$ is presented in Fig. 1 and can be approximated by the linear function:

$$N_0 = -8.81 + 0.564 \cdot P_s (\mathrm{MW\ s^{-1} \cdot cm^{-2}}). \qquad (2)$$

Eq. (2) gives a formal estimation for the laser ablation threshold: for the condition $N_0=0$, we get $P_s(min)=P_0=15.6$ MW cm^{-2}. At the power densities higher than 100 MW cm^{-2} Eq. (2) could be simplified: $N_0 \approx 0.564 \cdot P_s$.

Langmuir probes measurements. Velocities of the particle flow were estimated by time-of-flight measurements of cationic component of laser plasma. Measurements were carried out using two Langmuir probes connected through Koopmann circuit to digital scope TDS-360. Electrodes of the probes were fixed symmetrically with respect to the axis of the plasma flow. The time-of-flight of the cations t_i was determined at a fixed
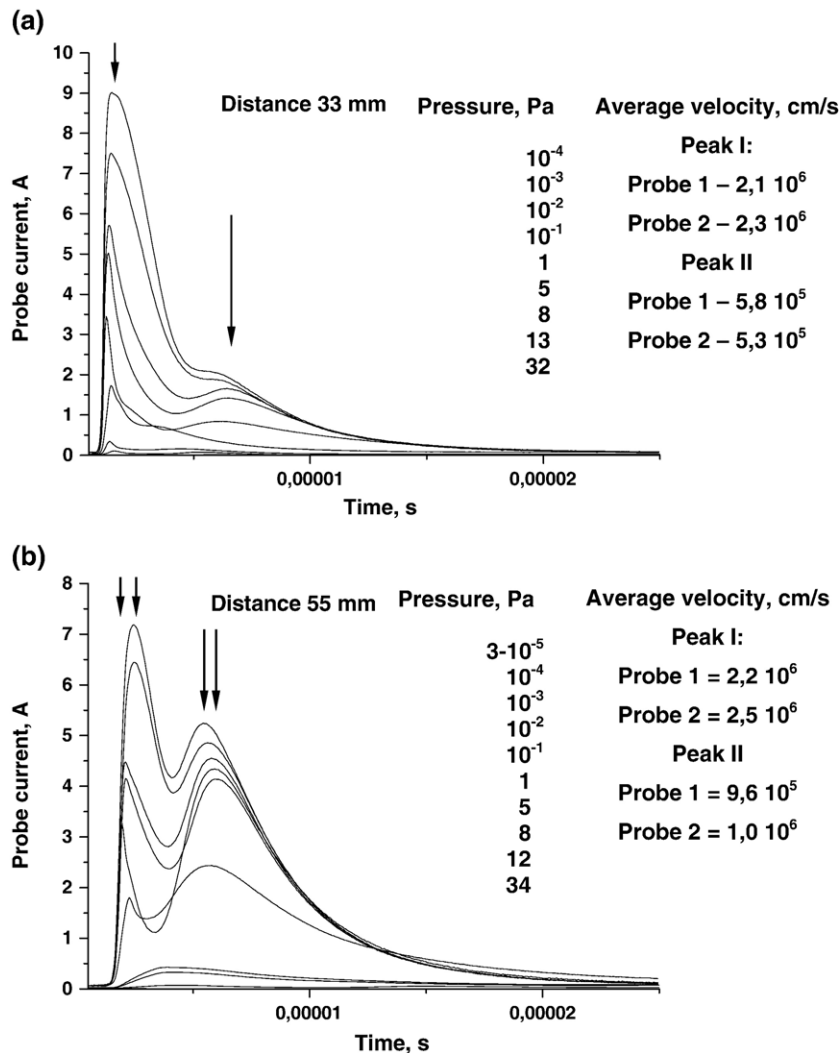

Fig. 2. Cationic current pulses measured at different oxygen pressures with a set of two Langmuir probes (bias voltage -15 V) at two fixed distances from the target: (a) -33 mm, (b) -55 mm.